HB 3/27/90
w/ ltr to attach #24

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H76

March 5, 1990

To: Members of the Marshall Islands Radiation Safety Review Committee

First, let me thank you for accepting Dr. Kato's invitation to participate in the Marshall Islands Radiation Safety Program Review. As you will see from the attached agenda and supporting materials, you will be reviewing all aspects of our work on this project.

You will note that the formal presentation will be completed on Monday with Tuesday reserved for your committee discussion and report preparation.

The committee will be composed of Roscoe Hall, who has graciously agreed to chair the meeting, Norman Cohen, Keith Eckerman, Henry Kohn, Leonard Newman, and Hylton Smith.

My wife and I plan to have you all (including accompanying persons) at our home for dinner on Monday evening.

The meeting will be held on March 26th and 27th in Room 2-22 of Building 130.

Sincerely,

CBM

Charles B. Meinhold
Division Head

CBM/gn
Enclosures
cc: J. Baum
H. Brown
J. Clinton
E. Kaplan
W. Kato
E. Lessard
A. Moorthy
G. Nubla
R. Pietrzak
W. Robison
J. Rudolph
C. Sun

Harry Brown NV

ENCLOSURES

1. AGENDA
2. Current Status of the Plutonium Level in the Rongelap and Utirik Urine Samples
3. The Radiological Dose from Pu at Rongelap Island
4. Marshall Islands Radiological Saftey Program Field Bioassay Mission 1989 Report
5. The Field Urine Collection Protocol
6. Derivation and Development of a Plutonium Fecal Excretion Function Using a Systemic Whole-Body Retention Function
7. Letter to Dr. Patricia Durbin
8. Letter to Mr. George Taylor
9. Whole Body Counting Daily Calibrations (DRAFT)
10. Exploratory Data Analysis (DRAFT)
11. Fission Track Analysis Quality Assurance
12. Plutonium from Atmospheric Weapens Testing: Fission Track Analysis of Urine Samples

Marshall Islands Radiological Safety Program
Review Agenda

Monday, March 26, 1990

Building 130 Conference Room

0830 - 0900	Welcome - Dr. W. Y. Kato
0900 - 0915	Introduction - Mr. Charles B. Meinhold
0915 - 0945	The Marshall Islands - Dr. C. Sun
0945 - 1030	FTA development and procedures/PERALS - Dr. A. Moorthy
1030 - 1045	Coffee Break
1045 - 1130	Urine collection and evaluation of FTA results - Dr. C. Sun
1130 - 1240	Lunch
1240 - 1350	Lab visit
1400 - 1530	Pu metabolic modeling - Dr. C. Sun
1530 - 1545	Coffee Break
1545 - 1600	Whole Body Counting system/calibration - Mr. J. Clinton
1600 - 1630	Whole Body Counting Quality Assurance - Dr. E. Kaplan
1630 - 1700	Discussion

Tuesday, March 27, 1990

0830 - 1200*	Executive Session
1200 - 1300	Lunch
1300 -	Close out with C. Meinhold and W. Kato

*Afternoon is available for extended discussion, if necessary.

Current Status of the Plutonium Level in the Rongelap and Utirik Urine Samples.

Executive Summary

At 6:45 a.m. the morning of March 1, 1954, a nuclear device, Code-named Bravo, was tested at Bikini Atoll. Unexpected weapon yield and tropospheric transport caused radioactive fallout to sweep over Rongelap and Utirik Atolls, 100 miles east from Bikini, a few hours later. As a result, thirty-five years following this incident, we are still studying the Northern Marshall Islands' radiological environments and evaluating the radiological impact on the Marshall Islands people.

As part of this effort, a comprehensive safety and dose reassessment project was conducted by Brookhaven National Laboratory (BNL) scientists beginning in 1981. Based both of the Lawrence Livermore Laboratory's (LLL) environmental measurements of air, water, food, and soil samples and the BNL's whole-body counting measurements, we presented a table of average annual effective dose equivalents (mrem/yr) from internal and external radiation (not including the dose from plutonium) to the people living at Rongelap Atoll. The total 30 years dose living on Rongelap Island was projected to be of less than 5,000 mrem. This is below the 170 mrem/yr of United States federal radiation protection guidelines for members of the public.

On May 1985, the people of Rongelap choose to leave their homeland and relocate on Majatto Island although the living conditions on Majatto were inferior to those on Rongelap. The basis for their relocation was never communicated to us, but it seem reasonable to assume that it may have been over their concern of plutonium in the environments taken from our polonium biased plutonium data in late 1984 from the Photon Electron Rejection Alpha Liquid Scintillation (PERALS) analytical methods.

In March 1, 1989, Dr. Kohn used the dose rate table mentioned above in his "Rongelap Reassessment Project Report." He showed that even using the 1987 maximum transuranic activity (5 fCi/sample) we found in urine, the estimated committed dose (i.e., the total dose to be received over the next 50 years), internal and external, from 1978 to 2008 still falls below an average of 170 mrem/yr.

As a result of our extensive evaluation of existing plutonium measurement for ultra-low activities in urine, a detection sensitivity of about 100 aCi/liter using fission track analytical (FTA) method was established at BNL in 1986. As of December 1988, over 500 urine samples collected from 1981 to 1984 from the Rongelap and Utirik people was completed. These measurements have met rigorous quality assurance standards for chemical analysis. However, some inconsistencies still existed in the FTA data which we presented during the Livermore meeting in February 1988.

Furthermore, all the 1988 urine samples (67 samples from the Rongelap people and 101 samples from the Utirik people) taken by Dr. Sun last September were just analyzed. The results support the thesis that soil contamination in some of the earlier urine samples was giving false information. Because of Dr. Sun's careful attention to collecting

uncontaminated urine samples, which was facilitated by Majatto's low soil concentration of plutonium, we were not surprised to find the statistics of our current Rongelap measurement reflect a median value far below the 250 aCi per sample as presented at the Livermore meeting.

Past studies of plutonium concentration in urine samples obtained from the Marshall Islands people have indicated levels much higher than those now known to be present. This is due, in part, to improved bioassay sample collection and analytical technology. Furthermore, from reanalyses of earlier samples and a more comprehensive review of the data, it now appears that earlier "high" plutonium results, were very likely due to: (1) naturally occurring polonium-210 inhaled in cigarette smoke and (2) by water and soil contamination of the urine samples during collection.

The polonium problem was resolved by the adaption of our FTA method. Regarding the soil contamination of the urine sample, the analysis of the September 1988 urine samples provided the following information:

1. From the samples taken in Majatto, all of the plutonium results are below 170 aCi (a committed effective dose equivalent 85 mrem). The median of the distribution is 27 aCi (a committed effective dose equivalent 14 mrem).

2. With one exception, the results from Utirik are similar to those of Rongelap. Including the one unverified high outcome, the median of the Utirik population is 24 aCi (a committed effective dose equivalent 12 mrem).

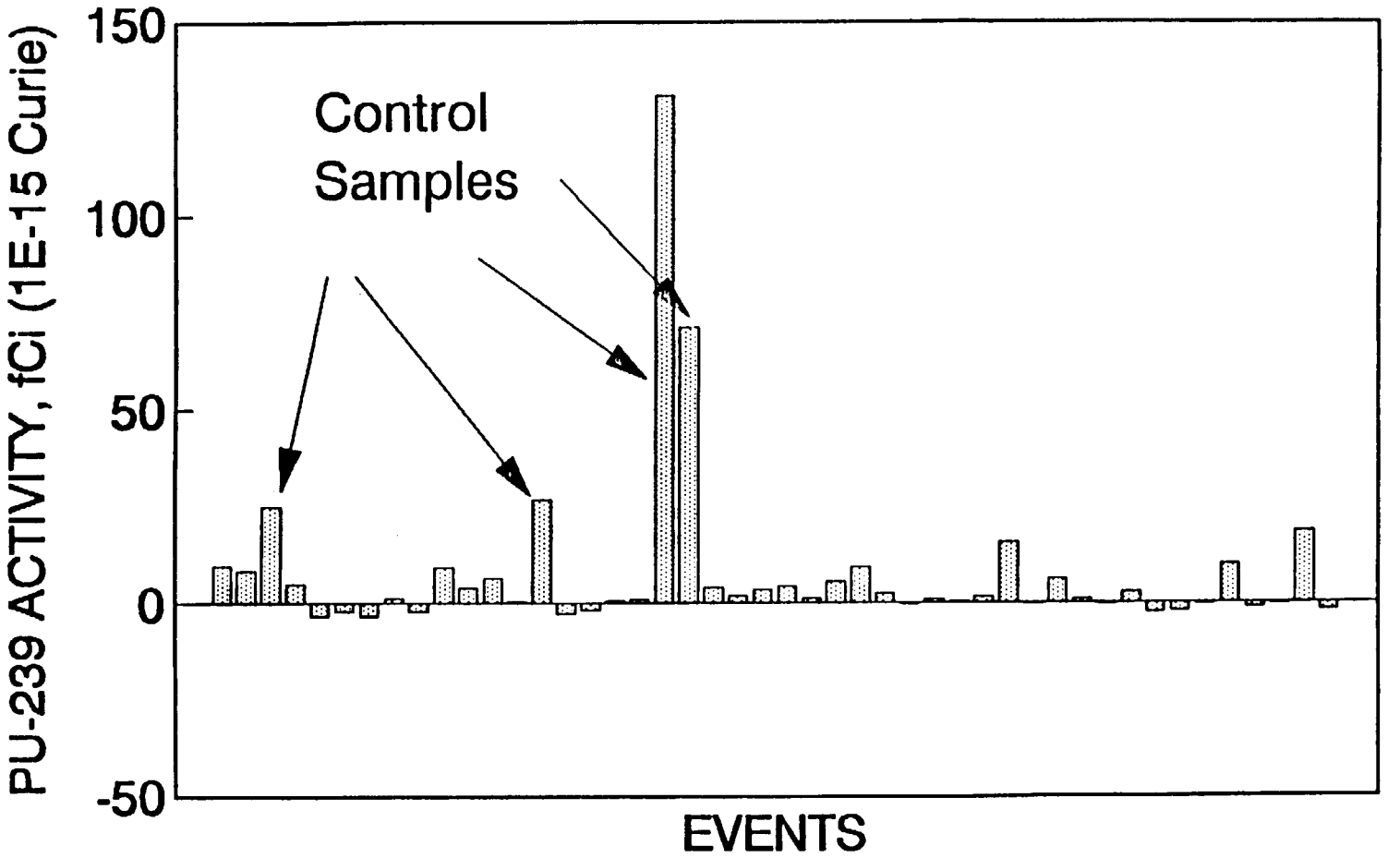
3. Statistical analyses indicate there are no differences between the mean and standard deviation of the distribution describing Rongelap's and Utirik's population at this time.

4. A most interesting observation is that the plutonium concentrations in the Islanders' urine samples is similar to that of our BNL individual which was used as our laboratory control up to December 31, 1988.

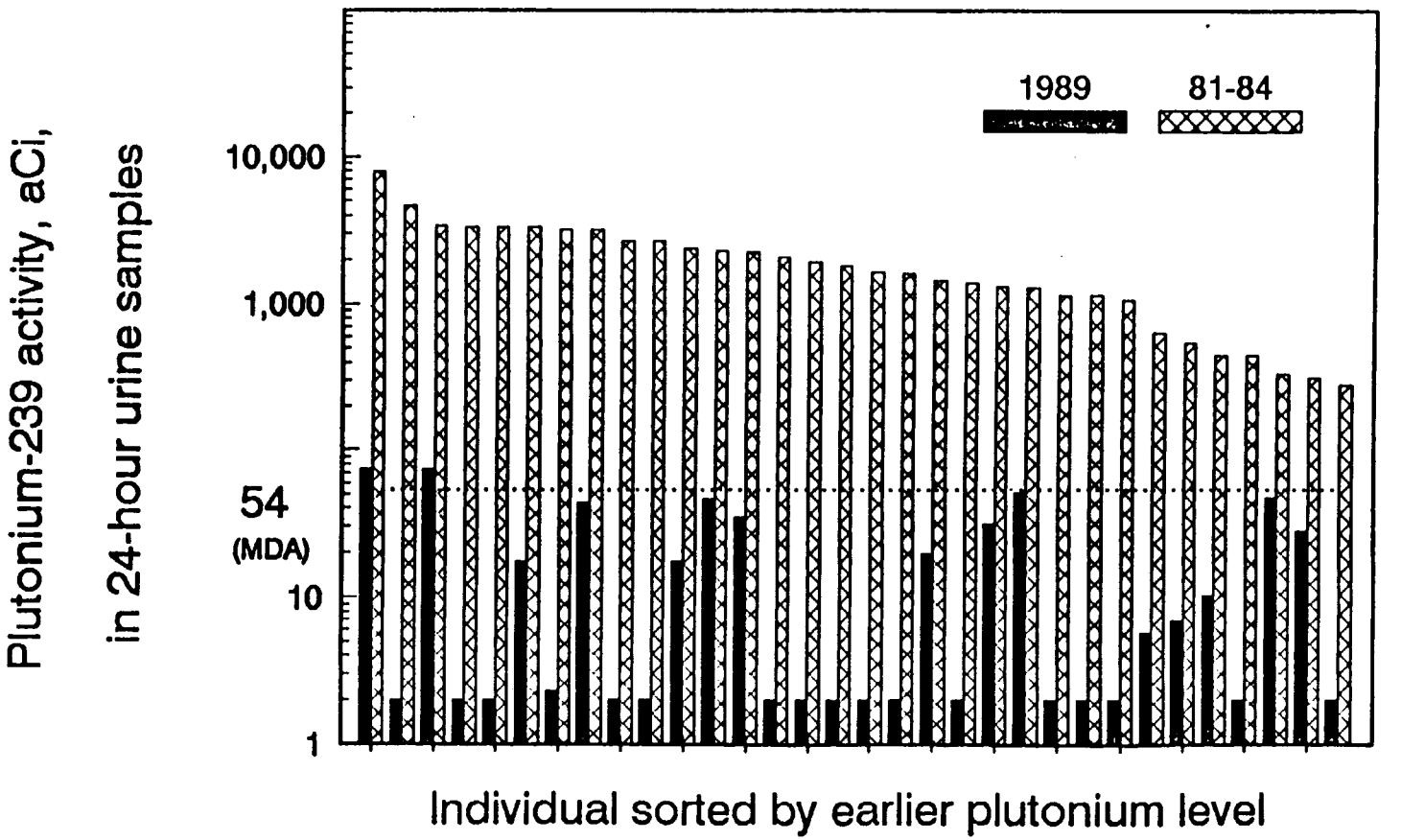
5. Using the maximum activity (200 aCi) and the most conservative retention model it would appear that all of the Islanders, but one, have a committed effective dose equivalent of less than 100 mrem (1 mSv). Even the one individual with the invalidated sample result mentioned above have a committed effective dose equivalent of less than 400 mrem (4 mSv).

With the greatly improved sensitivity of our FTA method and our newly developed urine sampling protocol we are confident that the Islanders' plutonium concerns can be satisfactorily answered.

MARSHALLESE URINE RESULTS
ANALYZED BY P.E.A.R.L.S.

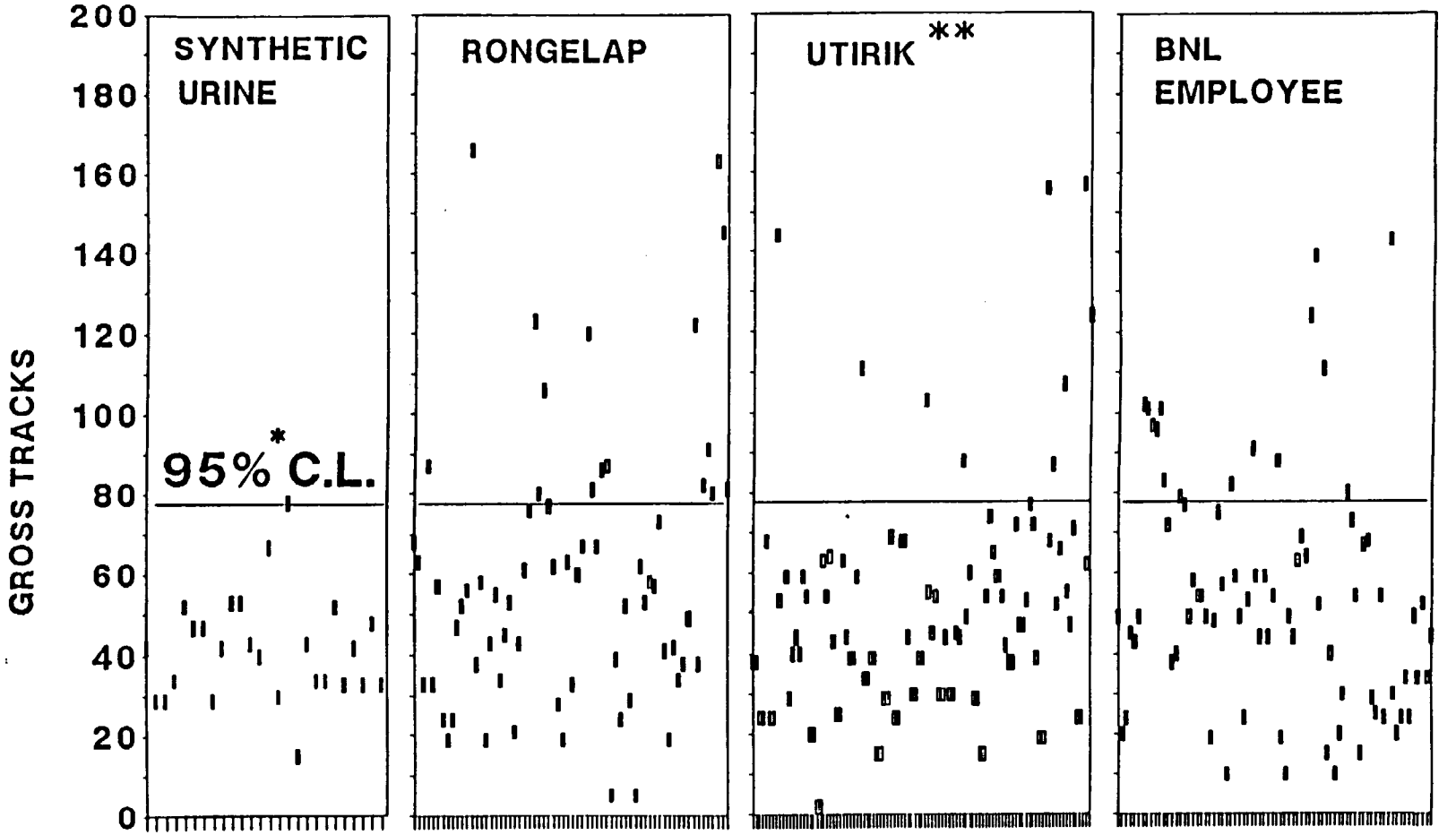


Comparison data of 32 resampled Islanders.



Note: for presentational purpose, 2 aCi is assigned as lowest value.

DISTRIBUTION OF URINE DATA
September 1988



* Data points above the 80 track line reflect samples which contain Pu at concentration greater than the synthetic urine at a 95% confidence level.

** One datum, 742 track, is not plotted. Because a 1981 sample was less then a MDL, new sample is being analyzed.

The Radiological Dose from Pu at Rongelap Island

W.L. Robison
C. Sun
C.B. Meinhold

A. Introduction

B. Dose Estimates
   1. Environmental Method (LLNL)
   2. Urine Analysis Method (BNL)
   3. Summary—Comparison Environmental and Urine Analysis

RONGELAP ISLAND

INTRODUCTION

The important issue to focus on when plutonium (Pu) and Americium (Am) are present in the environment is the potential radiological dose to people living in that environment. There are two basic methods for estimating this dose; one we will refer to as the "environmental method" and the other as the "urine analysis method." Other issues, such as the concentration of Pu in soil, are only relevant insofar as they provide information for the environmental method.

DOSE ESTIMATES

Environmental Method (Lawrence Livermore National Laboratory)

Pu in the soil is of no consequence if it is neither ingested nor inhaled. Thus, when Pu is present in the environment the potential radiological dose must be evaluated for both the inhalation and ingestion pathways. The radiological dose is dependent on the uptake of Pu by food crops and their subsequent ingestion by people, possible direct consumption of surface soil, and resuspension by wind of surface soil particles in the respirable size range that contain Pu which can be inhaled.

Uptake of Pu by food crops and resuspension of Pu contaminated surface soil are very dependent on environmental variables such as soil composition, soil pH, vegetation ground-cover, height of the vegetation canopy, and suspendability of the surface soil. If data are available for the uptake and resuspension of Pu for a specified environmental system, then these variables are accounted for and a direct and meaningful comparison can be made on the critical issue—the potential dose to people living in a specified environment.

We have analyzed many vegetation samples in the Marshall Islands, including Rongelap Island, to determine the concentration of Pu and Am in food crops. We find that plants have a very, very low uptake of Pu and Am and the consumption of soil is minor, being limited to occasional dust on ones hands. As a consequence, resuspension of plutonium contaminated surface soil, and the subsequent inhalation of Pu contaminated dust particles in the respirable

size-range, is the major potential route of exposure to people in the Marshall Islands as it is in almost any environment.

The resuspension of surface soil varies greatly, however, from one environment to another; resuspension may be very high in one environment and essentially negligible and of no consequence in another. Thus, it is much preferred that data for the concentration of Pu in air be available so that models can be developed relating Pu air concentration to Pu surface soil concentration, thereby eliminating much of the uncertainty in predicting resuspension mechanisms for a specific environment. We also have extensive data on the Pu and Am concentrations in surface soil and air from which we can estimate the amount of Pu and Am which might be inhaled or ingested during residence on Rongelap Island.

The 50-y integral effective dose equivalents for both the ingestion and inhalation pathways are based on the following:

Ingestion

1. The average concentration of Pu and Am measured in food products from Rongelap Island.

2. The ingestion of local foods based on the diet listed in Table A-1 of the attached Appendix A.

3. An assumption that 10 mg per day soil is ingested for every day of a person's life. We think this is conservative in that it overestimates the actual soil consumption of adults over their lifetime.

Inhalation

1. The average Pu concentration in air based on the LLNL resuspension model for Rongelap Island is conservatively estimated to be 190 aCi/m^3. This concentration is assumed to be present every day of a person's residence on Rongelap Island and when combined with the average breathing rate of 22 m^3/d gives the daily Pu inhalation rate in aCi/d. For comparison, the measured, average background

concentration of Pu in air at Bikini Island at Bikini Atoll and Enjebi Island at Enewetak Atoll, where the Pu concentration in the surface soil is 3 to 4 times higher than at Rongelap Island, is only about 30 to 60 aCi/m^3. Consequently, the average Pu concentration in air which we use to estimate the dose from inhalation is very conservative and, if anything, will overestimate the potential dose to people living on Rongelap Island.

2. The inhalation model as given in references 1 and 2.

The effective committed dose equivalent based on the above data is 75 mrem for Pu plus Am; the 50-y integral dose equivalent is 56 mrem. The relative contribution of Pu and Am and the inhalation and ingestion pathways is listed in Table 1.

To help put the estimated effective committed dose equivalent or the estimated 50-y integral effective dose from Pu and Am in perspective, we will compare them to the U.S. background dose. The average effective committed background dose equivalent in the United States is 300 mrem/y (3). Over 50 y this is a total effective committed dose of 15,000 mrem; the results are listed in Table 2. Based on our conservative estimates of the intake of Pu and Am by ingestion and inhalation, the estimated effective committed dose equivalent of 75 mrem due to Pu and Am at Rongelap Atoll is 200 times less than the average U.S. background dose over the same period of time.

The same conclusion, that Pu and Am at Rongelap contribute very minor radiation doses, can be reached by calculating an Annual Limit of Intake (ALI) for the general public from values listed in ICRP Publication 30 for radiation workers. An ALI for the public can be estimated by assuming that the ALI is a factor of 50 less than that for workers (5000 mrem divided by 50 equals 100 mrem). The results are shown in Table 3 and are converted from annual to daily intakes. The intakes at Rongelap for inhalation and ingestion are about 65 to 240 times less than one derives from the ICRP recommendations.

Table 1. The effective committed dose equivalent from Pu for 50 y of residence on Rongelap Island.[a]

	mrem		
	Inhalation	Ingestion	Total
Pu	34 (28)	12 (6.3)	46 (35)
Am	23 (18)	6.0 (3.4)	29 (21)
Total	57 (46)	18 (9.7)	75 (56)

[a] The 50-y integral dose equivalent is given in parentheses.

Table 2. The effective committed dose equivalent from Pu and Am at Rongelap Island and the effective committed background dose equivalent in the United States.[a]

	Effective committed dose equivalent, mrem[a]
Pu + Am dose at Rongelap	75 (56)
U.S. background	15,000

[a] The 50-y integral dose equivalent is given in parentheses.

Table 3. The annual intake of Pu via ingestion and inhalation at Rongelap Island compared with Annual Limit of Intake (ALI) for the public derived from recommendations by the ICRP for radiation workers. Intakes are converted from annual to daily intakes.

	Pu daily intake, pCi/d		
	Rongelap	ICRP (public)[a]	Ratio ICRP/Rongelap
Ingestion	0.18	44	244
Inhalation	0.0046	0.30	65

[a] Derived from ALI recommendations by ICRP for radiation workers (ICRP Publication 30, Part 4, 1988).

Urine Analysis Method (Brookhaven National Laboratory

In this method the Pu concentration in urine is determined by state-of-art fission track analytical (FTA) procedures. The measured Pu concentration is used in conjunction with excretion models for Pu to estimate the dose from Pu remaining in the body.

As of December 1988, over 500 urine samples collected during 1981 to 1984 from the Rongelap people were completed. Although these measurements have met rigorous quality assurance standards for chemical analysis, some inconsistencies still existed in the FTA data which we presented during the Livermore meeting in February 1988.

Now all 67 urine samples of the Rongelap people taken last September 1988 have been analyzed. The results support the thesis that soil contamination in some of the earlier urine samples was giving false information. Because of BNL's careful attention in September to collecting uncontaminated urine samples, which was facilitated by Majatto's low soil concentration of plutonium, we were not surprised to find the statistics of current Rongelap measurement reflect a median value far below the 250 aCi per sample as presented at the Livermore meeting.

Past studies of plutonium concentration in urine samples obtained from the Marshall Islands people indicated levels much higher than those now known to be present. The new sample data are, in part, the result of improved bioassay sample collection and analytical technology. Furthermore, it now appears that earlier "high" plutonium results were very likely due to: (1) naturally occurring polonium-210 inhaled in cigarette smoke and fresh fish and (2) water and soil contamination of the urine samples during collection.

The polonium problem was resolved by the adaption of our FTA method. Regarding soil contamination of the urine sample, the analyses of the September 1988 samples provided the following information:

1. From the samples taken in Majatto, all of the plutonium results are below 170 aCi (a committed effective dose equivalent 85 mrem, i.e., the total dose to be received over the next 50 years). The median of the distribution is at the background level.

2. An interesting observation is that the plutonium concentrations in the Rongelap people's urine samples is similar to that of our BNL individual who was used as our laboratory control up to December 31, 1988.

3. The mean Pu concentration in urine is below the FTA detection limit of 80 aCi; the 50-year effective committed dose equivalent based on the detection limit is about 40 mrem. The actual 50-year effective committed dose equivalent is something less than 40 mrem but how much less is unknown because of the detection limit.

SUMMARY

The radiological dose due to Pu in the environment at Rongelap is estimated by two very different methods (Environmental and Urine Analysis) and compared in Table 4.

The estimated effective committed dose equivalent (or the 50-y integral dose equivalent) due to Pu at Rongelap Island are very similar for the two quite independent methods. It is apparent that there is complete agreement between BNL and LLNL on the magnitude of the dose from Pu at Rongelap Island. Consequently, the 40 to 46 mrem effective committed dose equivalent (35 mrem 50-y integral dose equivalent) from Pu is insignificant when compared with the effective committed background dose of 15,000 mrem or more in the U.S. and other worldwide locations.

Table 4. The average effective committed dose equivalent from Pu at Rongelap Island in mrem.

	Method	
	Environmental (LLNL)	Urine Analysis (BNL)
	Effective committed dose equivalent	Effective committed dose equivalent
Pu	46 (35) mrem	40 mrem[a]
Am	29 (21) mrem	Assume Am 2/3 of Pu

[a] Based on the detection limit. The actual mean dose is something below this number.

REFERENCES

1. W.L. Robison, C.L. Conrado, and W.A. Phillips, Enjebi Island Dose Assessment, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53805 (1987).

2. J.H. Shinn, D.N. Homan, and W.L. Robison, Resuspension Studies at Bikini Atoll, Lawrence Livermore National Laboratory, Livermore, CA, UCID-18538 (1980).

3. National Council on Radiation Protection and Measurements, Exposure to the Population in the U.S. and Canada from Natural Background Radiation, National Council on Radiation Protection and Measurements, Washington, DC, NCRP-94 (1987).

APPENDIX A

Table A1. Model Diet: Kongelap Island. Local and Imported Food Available for Adult > 18 yrs.

Local Food	Grams/d	Kcal/g	Kcal/d	Specific Activity in 1990, in (pCi/g wet wt.) 137Cs	90Sr	239+240Pu	241Am	pCi/day 137Cs	90Sr	239+240Pu	241Am
fish	24.2	1.40	33.8	1.9E-02	6.5E-04	2.4E-04	4.2E-05	4.7E-01	1.6E-02	5.8E-03	1.0E-0
Tuna	13.9	1.40	19.4	1.9E-02	6.5E-04	2.4E-04	4.2E-05	2.7E-01	9.0E-03	3.3E-03	5.8E-0
Mahi Mahi	3.56	1.10	3.92	1.9E-02	6.5E-04	2.4E-04	4.2E-05	6.8E-02	2.3E-03	8.5E-04	1.5E-0
Marine Crabs	1.68	0.90	1.51	5.8E-04	1.6E-03	1.1E-03	1.9E-04	9.7E-04	2.7E-03	1.8E-03	3.2E-0
Lobster	3.88	0.90	3.49	5.8E-04	1.6E-03	1.1E-03	1.9E-04	2.2E-03	6.3E-03	4.2E-03	7.4E-0
Clams	4.56	0.80	3.65	1.2E-03	4.0E-03	1.0E-02	3.1E-03	5.6E-03	1.8E-02	4.6E-02	1.4E-0
Trochus	0.10	0.80	0.08	1.2E-03	4.0E-03	1.0E-02	3.1E-03	1.2E-04	4.0E-04	1.0E-03	3.1E-0
Tridacna Muscle	1.67	1.28	2.14	1.2E-03	4.0E-03	1.0E-02	3.1E-03	2.1E-03	6.8E-03	1.7E-02	5.2E-0
Jedrul	3.08	0.80	2.46	1.2E-03	4.0E-03	1.0E-02	3.1E-03	3.8E-03	1.2E-02	3.1E-02	9.7E-0
Coconut Crabs	3.13	0.70	2.19	2.7E+00	1.2E+00	1.9E-03	6.2E-04	8.5E+00	3.7E+00	6.1E-03	2.0E-0
Land Crabs	0.00	0.70	0.00	2.7E+00	1.2E+00	1.9E-03	6.2E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+0
Octopus	4.51	1.00	4.51	1.1E-02	1.6E-03	2.6E-04	4.6E-05	4.8E-02	7.3E-03	1.2E-03	2.1E-0
Turtle	4.34	0.89	3.86	3.1E-03	2.5E-04	8.2E-05	1.4E-05	1.3E-02	1.1E-03	3.5E-04	6.2E-0
Chicken Muscle	8.36	1.70	14.2	3.9E+00	4.1E-03	6.8E-05	9.0E-04	3.3E+01	3.4E-02	5.7E-04	7.5E-0
Chicken Liver	4.50	1.64	7.38	2.7E+00	8.7E-03	3.4E-04	8.4E-04	1.2E+01	3.9E-02	1.5E-03	3.8E-0
Chicken Gizzard	1.66	1.48	2.46	1.6E+00	9.7E-03	1.6E-04	2.7E-04	2.7E+00	1.6E-02	2.7E-04	4.5E-0
Pork Muscle	5.67	4.50	25.5	1.0E+01	2.7E-03	3.6E-05	2.5E-05	5.8E+01	1.5E-02	2.0E-04	1.4E-0
Pork Kidney	NR	1.40	0.00	1.3E+01	4.5E-03	3.4E-04	6.4E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+0
Pork Liver	2.60	2.41	6.27	5.7E+00	4.5E-03	9.1E-04	3.4E-04	1.5E+01	1.2E-02	2.4E-03	8.9E-0
Pork Heart	10.6	1.95	20.6	1.0E+01	2.7E-03	3.6E-05	2.5E-05	1.1E+02	2.9E-02	3.8E-04	2.6E-0
Bird Muscle	2.71	1.70	4.61	1.9E-02	6.5E-04	2.4E-04	4.2E-05	5.2E-02	1.8E-03	6.5E-04	1.1E-0
Bird Eggs	1.54	1.50	2.31	1.2E-02	2.9E-04	2.4E-04	4.2E-05	1.8E-02	4.5E-04	3.7E-04	6.5E-0
Chicken Eggs	7.25	1.63	11.8	3.9E+00	4.1E-03	6.8E-05	9.0E-04	2.9E+01	2.9E-02	4.9E-04	6.5E-0
Turtle Eggs	9.36	1.50	14.0	3.1E-03	2.5E-04	8.2E-05	1.4E-05	2.9E-02	2.3E-03	7.6E-04	1.3E-0
Pandanus Fruit	8.66	0.60	5.20	9.0E+00	4.4E-01	4.4E-05	2.2E-05	7.8E+01	3.8E+00	3.8E-04	1.9E-0
anus Nuts	0.50	2.66	1.33	9.0E+00	4.4E-01	4.4E-05	2.2E-05	4.5E+00	2.2E-01	2.2E-05	1.1E-0
Breadfruit	27.2	1.30	35.3	2.8E+00	6.1E-02	1.6E-05	2.0E-05	7.6E+01	1.7E+00	4.4E-04	5.4E-0
Coconut Juice	99.1	0.11	10.9	8.5E-01	1.1E-03	2.7E-05	2.5E-05	8.5E+01	1.1E-01	2.6E-03	2.5E-0
Coconut Milk	51.9	3.46	179	4.4E+00	1.6E-02	4.5E-05	5.5E-05	2.3E+02	8.2E-01	2.3E-03	2.9E-0
Tuba/Jekero	0.00	0.50	0.00	4.4E+00	1.6E-02	4.5E-05	5.5E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+0
Drinking Coco Meat	31.7	1.02	32.3	1.7E+00	1.0E-02	3.3E-05	3.8E-05	5.5E+01	3.2E-01	1.1E-03	1.2E-0
Copra Meat	12.2	4.14	50.3	4.4E+00	1.6E-02	4.5E-05	5.5E-05	5.4E+01	1.9E-01	5.5E-04	6.7E-0
Sprout. Coco	7.79	0.80	6.23	4.4E+00	1.6E-02	4.5E-05	5.5E-05	3.5E+01	1.2E-01	3.5E-04	4.3E-0
Marsh. Cake	11.7	3.36	39.2	4.4E+00	1.6E-02	4.5E-05	5.5E-05	5.2E+01	1.8E-01	5.2E-04	6.5E-0
Papaya	6.59	0.39	2.57	9.8E+00	1.4E-01	1.3E-04	6.2E-05	6.4E+01	9.2E-01	8.6E-04	4.1E-0
Squash	NR	0.47	0.00	6.3E+00	9.6E-02	1.7E-05	8.3E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+0
Pumpkin	1.24	0.30	0.37	6.3E+00	8.6E-02	1.7E-05	8.3E-06	7.8E+00	1.1E-01	2.1E-05	1.0E-0
Banana	0.02	0.88	0.02	1.1E+00	2.6E-02	1.3E-04	6.2E-05	2.2E-02	5.3E-04	2.6E-06	1.2E-0
Arrowroot	3.93	3.46	13.6	6.5E+00	7.8E-02	6.9E-04	3.6E-04	2.5E+01	3.0E-01	2.7E-03	1.4E-0
Citrus	0.10	0.49	0.05	1.8E+00	0.0E+00	0.0E+00	0.0E+00	1.8E-01	0.0E+00	0.0E+00	0.0E+0
Rainwater	313	0.00	0.00	5.2E-04	1.9E-04	2.4E-06	3.9E-07	1.6E-01	6.0E-02	7.5E-04	1.2E-0
Wellwater	207	0.00	0.00	8.0E-04	1.8E-03	1.3E-05	7.5E-06	1.7E-01	3.8E-01	2.7E-03	1.6E-0
Malolo	199	0.00	0.00	5.2E-04	1.9E-04	2.4E-06	3.9E-07	1.0E-01	3.8E-02	4.8E-04	7.8E-0
Coffee/Tea	228	0.00	0.00	5.2E-04	1.9E-04	2.4E-06	3.9E-07	1.2E-01	4.4E-02	5.5E-04	9.0E-0
Soil	0.01	0.00	0.00	1.6E+01	5.2E+00	4.0E+00	2.4E+00	1.6E-01	5.2E-02	4.0E-02	2.4E-0
Total Local	1332		567					1034	13	0.18	0.09
Fluids	1046		11								
Solids	286		556								

Marshall Islands Radiological Safety Program
Field Bioassay Mission 1989 Report
Brookhaven National Laboratory

SUMMARY:

From July 10, 1989, through August 12, 1989, under a contract with the United States Department of Energy (DOE), eight scientific team members from the Division of Radiological Sciences, Department of Nuclear Energy, Brookhaven National Laboratory (BNL), successfully completed a bioassay mission in the Republic of Marshall Islands. This bioassay mission included two major tasks: (1) to perform whole-body counting (WBC) measurements for quantifying the total amount of the measurable radionuclides in the body, and (2) to collect urine samples for evaluating plutonium level in the body. During this five week mission, a total of 976 WBC records and 209 urine samples were obtained from the people of Enewetak, Rongelap, Utirik and workers living in the Bikini field station.

The field organization and names of the eight members involved in this mission are shown in Attachment 1. The actual travel stops and schedule were followed as per the projected itinerary, Attachment 2.

All team members returned happy and in good health. This was a safe trip; there were no injuries or accidents. This was a productive trip; all team members adapted well to the sea and tropical weather and maintained daily duties. This was a successful program; we obtained urine samples from all, except one, of the individuals identified as providing a sample which indicated plutonium concentrations outside the normal distribution.

WHOLE-BODY COUNTING MEASUREMENT:

The purpose of this bioassay mission was to update WBC records for the people of Enewetak, Rongelap and Utirik and to reconfirm the radiological safety of the Islanders. The last Brookhaven WBC mission was conducted in 1985. Since then, the environmental conditions and radiological parameters may have changed, and the intake of residual fallout therefore needed to be determined.

The daily whole-body counting operation began at 0800 and ended about 1700. Two independent counting systems were operated to perform the whole-body counting measurements. The results were logged separately and the Islanders' data was tabulated into our database daily. The numbers of whole-body counts performed at each of the stops are summarized in next page:

	Marshallese Counted	Cross Count	Double Count	Non-Marshallese	Total Counts
Enewetak	217	5	7	6	235
Bikini	3	2	1	2	8
Majatto	91	5	4	4	104
Ebeye	142	5	2	2	151
Majuro	256	6	2	10	274
Utirik	197	4		3	204
Totals ==>	906	27	15	27	976

In the above table, the cross counts and double counts are used to determine the system checks and quality assurance of whole-body counting results. The cross count means an individual was counted twice by two different detectors. The double count means an individual was counted twice by the same detectors.

Following the past practice, a cover letter summarizing the WBC results and a set of preliminary whole-body data for the individuals counted were issued to the local leaders for each Island. In summary, during this five week survey, whole-body measurements did not reveal any unexpected activity. Cesium-137 and potassium-40 were the only radionuclides detected. Overall the average body burden of cesium in Islanders is declining. The average level of cesium varied from island to island, but the orders of magnitude are similar -- at approximately one ten-thousandths (1.E-4) of maximum permissible body burden (MPBB) of 30 uCi. However, individual cesium burdens varied greatly -- over three orders of magnitude. The highest activity we obtained is about 0.1 uCi among the Enewetak population (corresponding dose rate is about 15 mrem/year.)

Please notice that on this trip we used new whole-body counting equipment: new whole-body counting NaI(Tl) detectors, analyzers, computers and software. The physical properties and dimensions of the detectors are the same as the detectors used previously. While we enjoyed using this new whole-body counting systems, we also experienced problems during the mission; for example, overwriting data and the failure of analyzer performance due to unexpected electronic gain-shift.

All these operational problems will be evaluated and database of the whole-body counting from this trip will be finalized and released to DOE before January 1, 1990.

URINE COLLECTION PROGRAM:

Inconsistencies in the plutonium concentrations found in the urine samples collected from 1981-1984, raised the question of whether the urine collection and handling procedures in use at the time might have allowed soil/dust contamination of samples to occur. In order to prevent or minimize such contamination, a new urine collection method was developed for and used during

the present mission. Unlike the past, this time participants stayed aboard our vessel throughout the 24-hour collection period. Showers, clothing, meals and recreation (toys, games, audio and video tapes) were provided.

A maximum of 14 guests per day could be accommodated. The priorities were first to collect 24-hour urine samples from all persons found to be outliers in the earlier plutonium statistics, and second to obtain, whenever possible, repeated (more than once) 24-hour urine samples from these individuals. Again, a total of 209 urine samples was collected: 72 in Enewetak, 5 in Bikini, 33 in Majatto, 27 in Ebeye, 32 in Majuro and 40 in Utirik. This new bioassay sampling protocol and detailed daily working schedule are shown in Attachment 3.

To assure collection of contamination-free samples, the following procedures were used in the daily operation:

1. All participants were required to shower and change clothes at the beginning of each 24-hour period. Recreation and sleeping quarters were cleaned daily. These areas were off limits to the other islanders.

2. Urine collection participants wore yellow ID bracelets to differentiate them from WBC participants. These guests were restricted to the boat for their stay.

3. The time and volume of each urination by each participant were recorded by the nurse on duty. Any factors, such as bowel movements and menstruation, that may complicate later fission track analysis and interpretation, were also recorded.

4. Exit interviews were conducted to ensure that complete 24-hour urine samples were obtained properly from all participants. The most important aspect of this interview was that it allowed our nurse to review individual records and asks guests to give their last urine sample before they were discharged. Comments or feedback obtained during these interviews gave us very useful information to evaluate and improve our program.

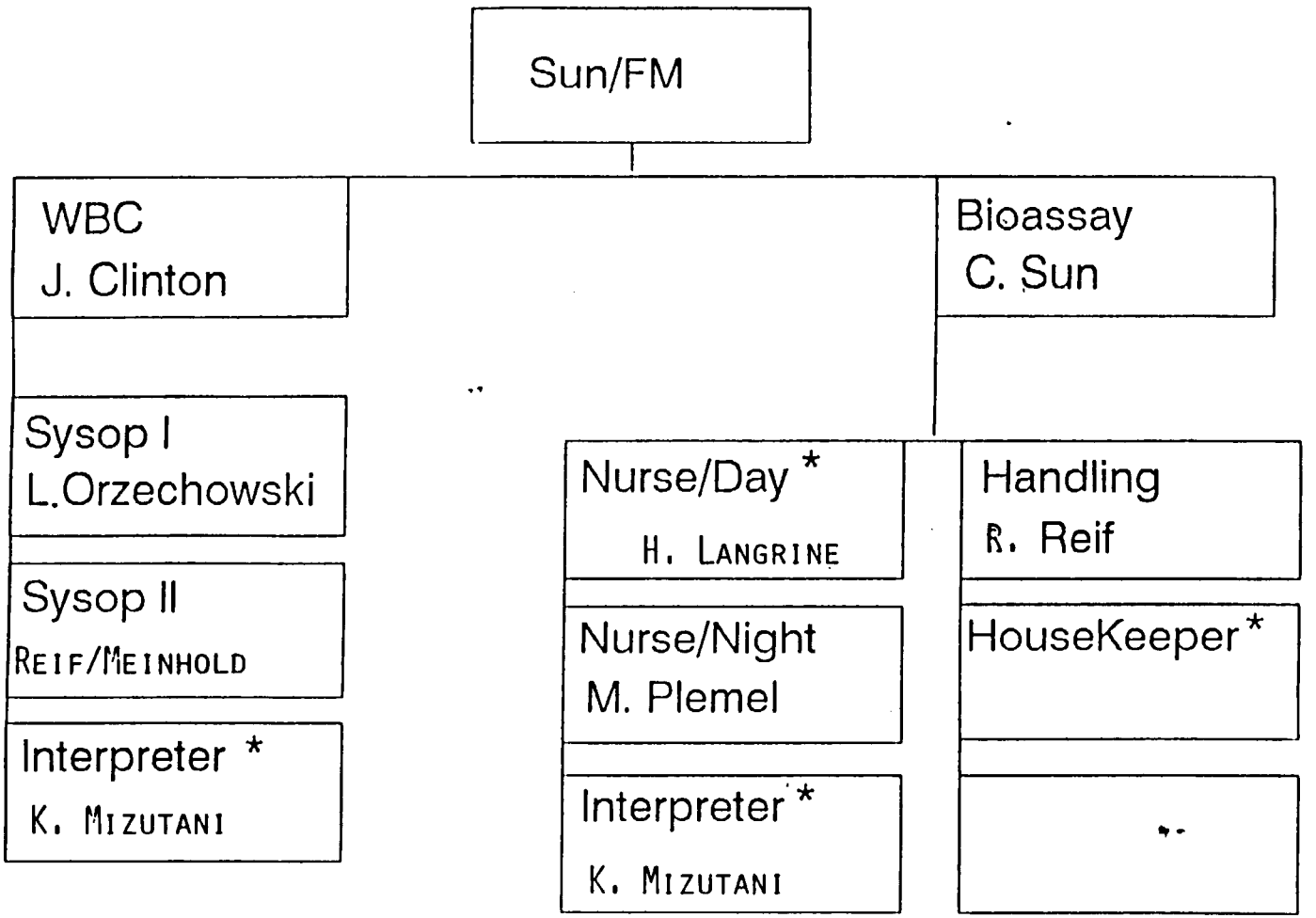
Because of the widespread low-level contamination in the Marshall Islands due to the nuclear tests in the islands, a true measure of the background level of plutonium due to nuclear testing elsewhere in the world cannot be obtained from the Marshallese themselves. Consequently, while we were at Majuro, a one-day, open invitation to non-Marshallese students at the College of Micronesia to participate in our bioassay program was posted on the college's bulletin board. Five students volunteered; 3 from Pohnpei and 2 from Truk. The results of these five samples may provide an indication of the contribution of worldwide nuclear testing to the total of plutonium burden measured among the Marshallese.

Five 24-hour urine samples were obtained from the workers living at the Bikini Field Station, Bikini Island. Aside from their value for the radiation monitoring of the worker, the urinalysis results of these five samples could provide a small set of human data on the plutonium uptake by chronic

ingestion. Such information might be useful for considering settlement options and projecting dose for the people of Bikini after their return.

The urine samples from the people of Rongelap who had plutonium greater than 1,000 aCi in earlier samples are presently being processed. The target date for release of these urine results is January 1, 1990. Finally, we thank Mr. Reynolds DeBrum, Dave Wheeler, Bill Jackson, John Brown, H&N participants and G. W. Pierce crew members for making this mission successful.

1989 Summer Orgnization



* Marshallese Staffs

B - AT ANCHOR, BIKINI
E - AT ANCHOR, ENEWETAK
EB - AT DOCK, EBEYE
K - AT DOCK, KWAJALEIN
M - AT DOCK, MAJURO
MJ - AT ANCHOR, MEJATTO
T - TRANSIT
P - AT ANCHOR, RONGELAP
U - AT ANCHOR, UTERIK

MISSIONS

- Resupply Enewetak/Bikini
- Runit Dome Survey
- Whole Body Count (WBC)
- U.S. Navy-Nat'l Pers Service-File Crew for Ships Survey
- Mini-Medical Mission

VESSEL SCHEDULE
G.W. PIERCE

	tue	wed	thu	fri	sat	sun	mon	tue	wed	thu	fri	sat	sun	mon	tue	wed	thu	fri	sat	sun	mon	tue	wed	thu	fri	sat	sun	mon	tue	wed	thu	fri	sat	sun
			1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30		
june			K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K/T	T/B	B	B	B	B	B/T	T/E	E/T	
																									BIK/ENWTK RESUPPLY/RUNIT DOME SURVEY									
					1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30
july			T/K	K	K	K	K	K	K	E/T	T/E	E	E	E	E	E	E	E/T	T/B	B/T	T/MJ	MJ	MJ	MJ/T	T/K	K/EB	EB	EB	EB	EB/K	K/T	T/M	M
																									WBC MISSION									
									1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26
august	M	M	M	M	M/T	T/U	U	U	U	U	U	U/T	T/K	K	K	K	K	K/T	T/B	B	B	B	B	B	B	B	B	B	B	B	B	B	B	B
																									WBC MISSION									
																									NAVY-NPS SHIPS SURVEY									
september	B	B	B	B	B/T	T/K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K	K/MJ	MJ	MJ	MJ	MJ	MJ	MJ	MJ	MJ	MJ	MJ	MJ	MJ/T
																												MEDICAL MISSION						

NAVY-NPS SHIPS SURVEY

DAILY SCHEDULE FOR PARTICIPANTS UNDERGOING URINE SAMPLING

1. **6:00 pm - 7:00 pm** **Dinner for crew only**

   Overnight participants (16) arrive ... snacks/beverages available to them ... each is given soap and a towel, allowed to shower, and are provided with tee shirts and shorts (for men) or moo-moos (for women).

2. **8:00 pm - 8:30 pm** Marshallese nurse instructs participants in urine collection procedures. (Note: as described in the Urine Collection Procedure documentation, it is the nurses' responsibility to assign urine collectors as necessary, to decant samples into each individual's larger sample bottle, and to mark on this sample bottle each individual's ID#, name. Nurses will also keep a detailed log including the time, volume, id#, and name for each INDIVIDUAL sample.)

3. **8:30 pm** Free time: videos, movies, games, entertainment.

4. As early as possible each morning: It is the on-duty nurse's responsibility TO ENSURE THE TAKING THE FIRST MORNING URINE SAMPLE FROM EACH PATIENT! This is the most important sample of all.

5. **6:30 am - 8:00 am** Breakfast for crew and participants.

6. **7:30 am - 10:00 am** Begin whole body counting on the 16 individuals who remained onboard the previous evening.

7. **10:00 am - 12:00 pm** Free time for previous evenings' participants.

   New participants for whole body counting start arriving ... WBC process for these 54 ADDITIONAL individuals takes approximately 7-9 hours (est 15-20 minutes per individual, repectively, with two WBC chairs in use simultaneously).

8. **12:00 pm - 4:00 pm** Free period for previous evenings' participants during which time they are given exit interviews

9. **4:00 pm** Return participants to island. They will be allowed to keep the clothes issued to them.

THE FIELD URINE COLLECTION PROTOCOL

MARSHALL ISLANDS RADIOLOGICAL SAFETY PROGRAM

BROOKHAVEN NATIONAL LABORATORY

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INTRODUCTION

Inconsistencies in plutonium concentrations found in Marshallese urine samples collected in 1981 to 1984 suggested that urine collection and handling procedures in use at the time resulted in soil and/or dust contamination. New urine collection methods, which prevent or minimize the possibility for such contamination, were developed and used in the summer of 1989 during our most recent Marshall Islands bioassay mission. For the first time, participants were required to stay aboard the mission vessel throughout the 24-h collection period. In addition, each participant was required to shower and change into clean clothing. These requirements were judged essential in meeting the goal of collecting contamination-free 24-h samples.

The Brookhaven National Laboratory (BNL) Marshall Islands Radiological Safety Program, which operates under contract to the Department of Energy, has been active since 1978. Today, the major tasks of the program are:

1) developing adequate bioassay systems to measure through either urine/fecal analysis and whole-body counting the low levels of radioactivity in the Islanders' bodies and 2) comparing these results against the predicted values provided by the Livermore National Laboratory based on their environmental analysis. In this way, BNL can help ensure that the exposure of the Islanders is well within the existing guidelines.

From 10 July 1989 through 12 August 1989, the Brookhaven team conducted its most recent field trip in the Republic of the Marshall Islands. This mission included two major tasks: 1) whole-body counting measurements for quantifying the total amount of radionuclides emitting penetrating radiation in the bodies of the Marshall Islanders and 2) urine sample collection for evaluating plutonium levels in these same people.

Questions have been raised about results of plutonium measurements from previous missions. One possible explanation of the measured high values is that these samples were contaminated, probably by airborne soil particles. Because of such potential problems, a totally new technique was developed for the measurement of ultra-low plutonium activity in urine (Moorthy et al. 1988). A reliable detection sensitivity of about 3.7 μBq using this new fission track analytical (FTA) method was established at BNL in 1986. As of December 1988, over 600 plutonium measurements were made using the FTA method from urine samples collected during 1981 to 1984 from the Rongelap and Utirik people. These measurements met rigorous quality assurance standards for chemical analyses, but some inconsistencies still existed. These have now been overcome, and the FTA method has a reliable detection sensitivity of about 2 μBq.

Since a major objective of this mission was to test the validity of the hypothesis that the "higher" plutonium concentrations in some of the urine samples were due to soil contamination during collection, the following extraordinary measures were instituted:

1. At the beginning of each 24-h period all participants were required to shower and change into clean clothes provided by the staff. Recreation and sleeping quarters were thoroughly cleaned daily. Entrance into these areas was restricted to participants and staff.

2. Urine collection participants were issued yellow ID bracelets to differentiate them from those undergoing whole-body counting since only the urine collection participants were restricted to the boat for 24 h.

3. The supervising nurse recorded the volume and time of each urination. Other factors that could have complicated later fission track analysis and interpretation, such as bowel movements and menstruation, were also recorded.

4. Exit interviews were conducted with all participants. The most important aspect of this interview was that it allowed the nurse to review individual records and ask participants to give their last urine sample before they left. Comments obtained during these interviews provided useful information to evaluate and improve the collection program.

During this mission only 14 participants per day could be accommodated because of limited space and staff. Priorities, therefore, had to focus first on collecting urine samples from all persons for whom previous plutonium analysis measurements were found to be outside the normal range, and second on obtaining whenever possible repeated 24-h samples from these individuals. It was only after these requirements were met that the collection of samples from the remaining Rongelap people could be addressed. The data recorded on urine volume showed that 24-h urine production among the Marshallese, regardless of age or sex, varied from 50 to 3,500 mL. The large volume samples might be explained by the large volume of soft drinks ingested by the participants during their stay. The lower end of the range of volumes is somewhat more difficult to understand although individuals living in hot environments tend to have urine elimination volumes far below those of reference man. Even under these assumptions, however, it would seem unreasonable to expect that a 50 or 100 mL sample represented a true 24-h elimination. Figure 1 depicts the volume distribution of 209 urine samples obtained during the summer of 1989. The mean and standard deviations are approximately 950 mL and 450 mL, respectively.

Taking the entire Marshall Islands population resampled as a whole, the mean urine volume value of the 209 urine samples was close to the elimination value given for reference man by the International Commission on Radiological Protection (1975). However, it is clear that taking the ratio of the 24-h collection samples to a standard man elimination volume could result in extremely unrealistic estimates of body burden and intake. It is important to recognize that for the interpretation of individual's urine data, it is the collection period, not the volume, that is the parameter of importance.

The first measurements from the 1989 mission are from samples collected from individuals with the highest plutonium activities found from previous missions (1981-1984). This information has just become available and is shown in Table 1 and plotted in Fig. 2. In both places, 75 nBq was assigned as the lowest value for those measurements 75 nBq or less for presentational purpose. Using the FTA method, it seem that only two individuals have plutonium activities above the new Minimum Detectible Amount (MDA) of 2 μBq per sample. At least in these individuals, the new protocol has substantially reduced the possibility of soil contamination during the collection of samples. These Islanders will be resampled during the next mission to confirm these results. As an aside, it is noted that 3.7 μBq represents a dose rate of about 1 mrem per year.

The entire 209 urine samples collected on the mission will be analyzed by the FTA over the next year. They will be compared with data taken previously to confirm or reject the soil contamination hypothesis. Hopefully, the use of the explicit and strict sample collection procedures outlined above will remove at least this question from acceptance of the data reported on the Marshall Islanders. A full report on results of the 1989 mission will be published as quickly as possible.

This work was performed under the auspices of the U.S. Department of Energy under contract DE-AC02-76CH00016.

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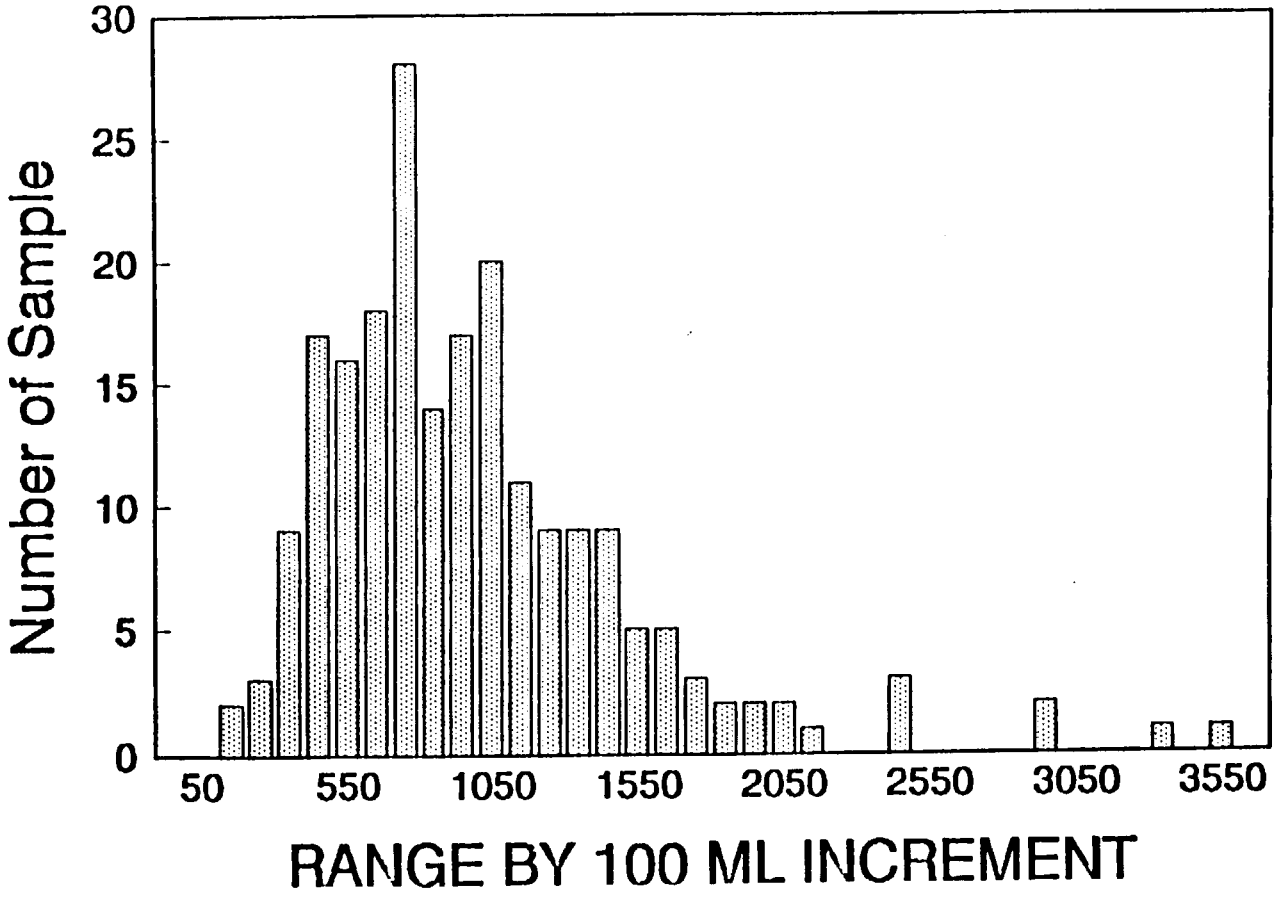

Figure 1. Volume Distribution of 209 Urine Samples

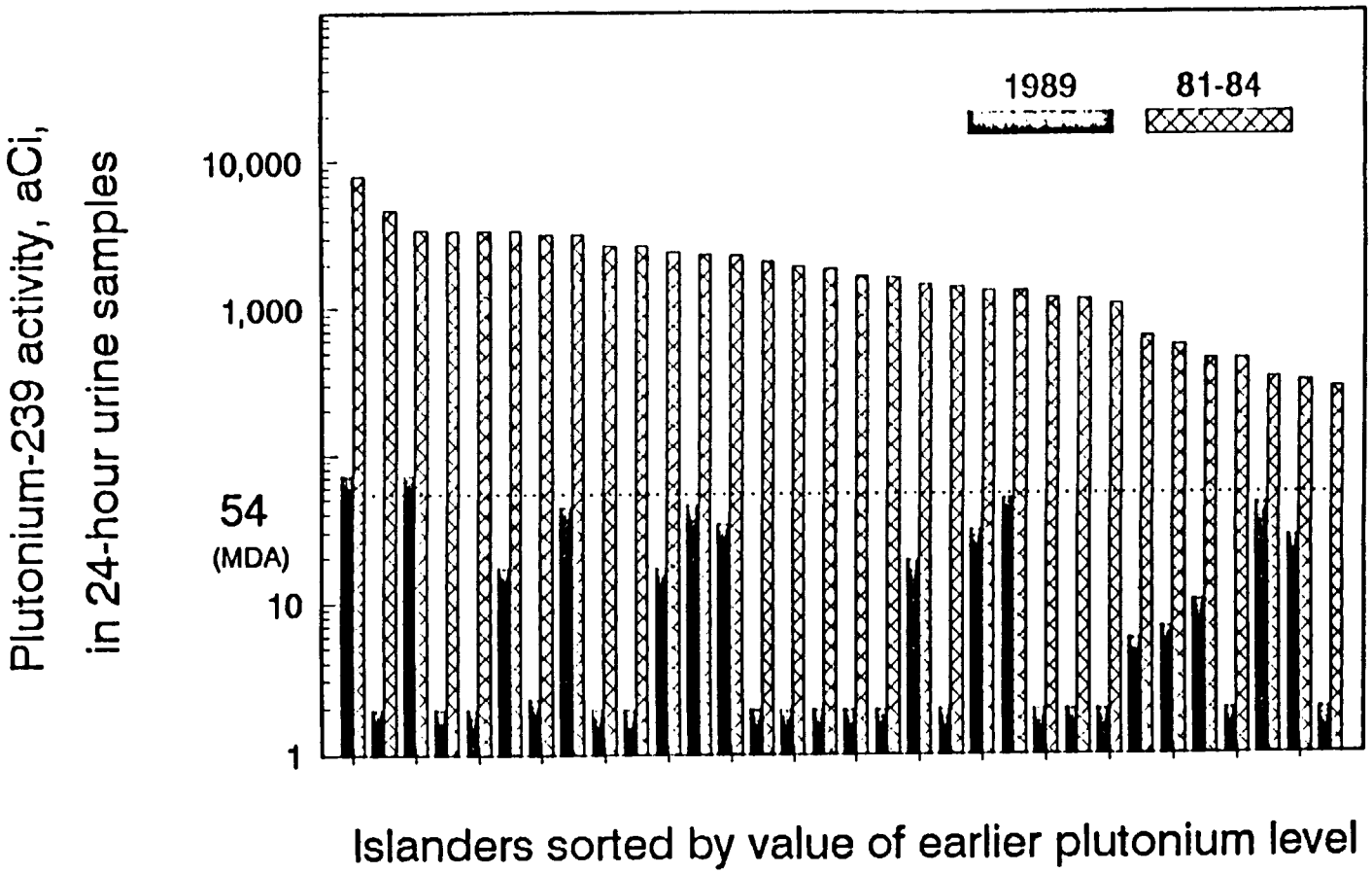

Figure 2. Comparison data of 32 resampled individuals.

Note: for presentational purpose, 2 aCi is assigned as lowest value.

Table 1. Plutonium-239 urine data from 32 samples.

Sample #	Fission track	1989 data Pu-239 (aCi)‡	81-84 data Pu-239 (aCi)
1	102	74	8025
2	25	2§	4722
3	102	74	3442
4	34	2	3390
5	34	2	3390
6	53	17	3390
7	40	2	3220
8	76	44	3220
9	23	2	2690
10	15	2	2690
11	53	17	2425
12	78	46	2335
13	68	35	2296
14	9	2	2089
15	29	2	1938
16	34	2	1853
17	34	2	1662
18	34	2	1630
19	55	20	1452
20	29	2	1386
21	65	31	1308
22	82	51	1295
23	34	2	1151
24	38	2	1145
25	24	2	1055
26	43	6	638
27	44	7	549
28	47	10	448
29	33	2	448
30	79	48	333
31	62	28	316
32	34	2	282

‡: Values less than 54 aCi is below current MDA.
§: 2 aCi is assigned as lowest value for those 2 aCi or less.

later reviewed and challenged by others (Beach and Dolphin 1964; Gerber et al. 1989; Stover and Jee 1972; ICRP 1986). Based on examination of the fecal Pu excretion in relationship to the patient's medical status, Durbin suggested instead that in a healthy individual Pu excretion in feces is slightly higher than Pu excretion in urine after an acute uptake, administrated by injection into the blood (Durbin 1972). Durbin reported that by the end of the second week after such an uptake the amount of Pu excreted in a 24-h urine sample was approximately equal to that in feces and the U:F ratio increases to 1.5 at about 100-d (Durbin 1972).

In addition to Langham's U:F ratio, Fig. 1 also presents the U:F ratio calculated using the model proposed in this paper. This ratio (hereafter referred to as the Sun U:F ratio) varies over three orders of magnitude within a 30-y period from a single Pu uptake. Due to the scarcity of human data relating to Pu excretion in feces, the Sun U:F predictions remain to be verified. However, the model could be used to derive a fecal excretion function from available systemic whole-body retention functions. The mathematics routine provided in this paper can be used for evaluating the consistency between existing Pu retention and excretion models.

A Generalized Mammillary Model

A mammillary model is a complex multi-compartmental model designed to represent a human or animal anatomical configuration. The model is used to describe transport phenomena of a contaminant among compartments representing body organs and tissues. Therefore, such a model can be used to study contaminant distribution and excretion in the body.

a most generalized anatomic configuration of human and which incorporates known urinary and fecal excretion pathways is developed. Using this model, the transport phenomena of the contaminants in the system, including the fecal excretion function, can be determined without kinetic information. This requires only an exponential systemic-whole body retention function or a urinary excretion function. The mathematical methods and computational algorithm for the model are also established and applied to plutonium metabolic models.

This study introduces: (1) A physiological model for the development of a fecal excretion function; (2) An analytical routine for identifying the translocation parameter values; (3) A method for calculating annual/committed organ dose from a systemic whole-body retention function.

INTRODUCTION

At early times after an intake excretion data obtained from fecal specimens can be as important as information obtained from 24-h urine samples for the estimation of dose from uptakes of plutonium (Pu). In Langham's human experiments in 1945 Pu was intravenously administered to individuals after which their daily urine and fecal specimens were analyzed over a 138-d interval (Langham 1956). These results showed that the Pu content in feces was about three times more than that in 24-h urine samples for the first few days. The ratio of Pu in urine to that in feces (U:F) was found to vary from 0.37 to 10 over a 30-y interval (Fig. 1). Langham's experimental data were

DERIVATION AND DEVELOPMENT OF A PLUTONIUM FECAL EXCRETION FUNCTION USING A SYSTEMIC WHOLE-BODY RETENTION FUNCTION

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ABSTRACT

Liver biliary secretion influences the radioactive contaminant content in feces. To enhance reliability of metabolic models and further increase the interpretation accuracy of bioassay data, a model that simulates

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This work was performed under the auspices of the U.S. Department of Energy under Contract No. DE-AC02-Ch00016.

In theory, a mammillary model consists of one central compartment and several peripheral compartments (Jacquez 1985). The central compartment is often identified with blood plasma and/or extracellular fluid. The number of peripheral compartments depends on the physical and chemical properties of the contaminant, i.e., where it is retained in terms of partitioning within physiological organs and tissues, (e.g., liver, bone, soft tissues, etc.). Due to the vascular structure, the contaminant elements are transported with blood bidirectionally throughout the entire body. Each peripheral compartment has a set of input and output pathways connected to the central compartment. These peripheral compartments are therefore connected to one anther only indirectly via the central compartment. However, if applicable, peripheral compartments can have an excretion pathway.

Mathematical Equations of the Generalized Mammillary Model

Fig. 2 presents the generalized mammillary model with M arbitrary compartments, consisting of one central compartment and M-1 peripheral compartments. Each compartment has its own excretion pathway.

For mathematical simplification, assume that the contaminants entering all compartments are uniformly mixed and are distributed instantaneously. Also assume that the transport and excretion of the contaminant are governed by a first order kinetics (Levenspiel 1972). That is, the instantaneous rate of changed atoms in a volume is a multiple product of the number of total atoms in the volume and a specific translocation rate constant (TRC) of unit reciprocal time.

Let $\kappa_{1,1}$ be a description of one TRC value that describes the instantaneous fraction of contaminant transport per unit time from the central pool (Compartment Number 1) to peripheral compartment i. Then, $\kappa_{1,i}$ is the TRC that is opposite in terms of direction to $\kappa_{i,1}$. Let $\mathfrak{R}_n(t)$ describe a compartmental retention function (CRF) for the n^{th} compartment of interest. The function gives fractions of an uptake presented in the compartment as a function of time t; where n is a numerical index for specifying a compartment (n=1 is reserved for the central compartment).

Assuming all contaminant is injected once into the central compartment at a reference time t=0, the initial conditions are: (1) The sum of the coefficients in the central compartment retention function (CCRF) $\mathfrak{R}_1$ should be unity. (2) The sum of the coefficients in all peripheral CRFs, $\mathfrak{R}_2$, $\mathfrak{R}_3$, ..., and $\mathfrak{R}_m$, are zero. The first-order differential equation for the contaminant in the central compartment for the model shown in Fig. 2 is:

$$\mathfrak{R}'_1(t) = \sum_{i=2}^{M} \kappa_{1,i}\, \mathfrak{R}_i(t) - k_1\, \mathfrak{R}_1(t) + \delta(0), \qquad (1)$$

Equation 1 is developed by writing a mass balance over a volume of the central compartment based on the equation of continuity for an isothermal system (Bird et al. 1960), i.e., the rate of mass accumulation in a compartment shall be equal to the rate of mass input less the rate of mass output. Likewise, the differential equations for the contaminant in the peripheral compartments are:

$$\mathfrak{R}'_2(t) = \kappa_{2,1}\, \mathfrak{R}_1(t) - k_2\, \mathfrak{R}_2(t), \qquad (2)$$

$$\mathfrak{R}'_3(t) = \kappa_{3,1}\, \mathfrak{R}_1(t) - k_3\, \mathfrak{R}_3(t), \text{ and} \qquad (3)$$

$$\pi'_m(t) = \kappa_{m,1}\,\pi_1(t) - k_m\,\pi_m(t). \tag{4}$$

where the $\pi'_n(t) = d\pi_n/dt$ and the $\delta(0)$ represents a delta function to describe a unit pulse input at time t=0 and the k_i's are the total removal rate constants. The total removal rate constants, k_1, k_2,..., k_m are defined:

$$k_1 = \beta_1 + \kappa_{2,1} + \kappa_{3,1} + \ldots + \kappa_{m,1}$$

$$= \beta_1 + \sum_{i=2}^{M} \kappa_{i,1} \tag{5}$$

$$k_2 = \beta_2 + \kappa_{1,2}, \tag{6}$$

$$k_3 = \beta_3 + \kappa_{1,3}, \ldots, \text{ and} \tag{7}$$

$$k_m = \beta_m + \kappa_{1,m}. \tag{8}$$

The β'_js are also a part of the TRC constants. Unlike the $\kappa_{i,j}$s, which are used for allowing recycling of contaminant between central and peripheral compartments, the β'_js are used for allowing excretion of contaminant from each of the compartments directly.

Inverse Problems and Degrees of Freedom

With a fully specified set of TRC values, eqns 1 through 8 can be easily solved. The analytical solution is a set of M-term exponential functions, π_1, π_2, π_3, ..., and π_m. Because each of these CRFs pertain to an identical set of eigenvalues, they can be summed to obtain a function R(t) as following:

$$R(t) = \sum_{n=1}^{N} \pi_n(t) = \sum_{n=1}^{N} \sum_{j=1}^{M} \alpha_{j,n} \, Exp(-\gamma_j t), \qquad (9)$$

where, $\alpha_{j,n}$'s are the constant coefficients and γ_j's are the rate constants with a reciprocal time unit. The n and j are two numerical indexes: The j is for the number of exponential terms expressed in a CRF and n is for the number of compartments in the model. The R(t) is an analytical sum of all CRFs; therefore, it can be called the systemic whole-body retention function (SWBRF).

The $\alpha_{j,n}$'s and γ_j's values needed in eqn 9 can be easily solved from eqns 1 through 8 for a specific mammillary model with a set of finite TRC parameter values. The techniques that are associated with obtaining the TRC ($\kappa_{1,j}$ and β_j) parameter values by using eqn 9 ($\alpha_{j,n}$ and γ_j) is called the inverse problem (Jacquez 1985). Inverse problem is not always difficulty to solve if the number of independent equations and the number of independent variables are equal. If the number of variables is greater than the number of equations, there will be no unique solution; if fewer independent variables are fixed, an infinite number of solutions will exist.

PROPOSED HEPATIC MODEL

Figure 3 shows a proposed model with a total of four compartments. This proposed model is simplified from the generalized mammillary model as shown in Fig. 2. The number of compartments in Fig. 3 can be increased or

decreased according to its application, but the number of direct excretion pathways shall always be two: β_1 in the central compartment (Compartment No. 1) and β_2 in one of the peripheral compartments (Compartment No.2). Because the proposed model incorporates β_2 to simulate the liver-bile secretion pathway, the model can be used to evaluate the direct excretion of contaminant from the liver compartment to feces. Therefore, the model is named the Hepatic model to reflect this property.

Transfer Functions

Let $Z_n(S)$ be the transfer function of a retention function $\pi_n(t)$ yielded by the Laplace transformation. First, take the Laplace transform of eqns 1 through 4. Second, substitute the Laplace functions of eqns 2 through 4 into the Laplace function of eqn 1. Then, the transfer function for the central compartment $Z_1(S)$ can be organized as:

$$Z_1(S) = \frac{\prod_{j=2}^{M} (S + k_j)}{\{ S+k_1 - \sum_{n=2}^{M} \frac{\rho_n}{(S+k_n)} \} \prod_{j=2}^{M} (S+k_j)}, \qquad (10)$$

where $\rho_n = \kappa_{1,n} \times \kappa_{n,1}$, i.e., the product of the pair of TRCs representing translocation between the central compartment and an n^{th} peripheral compartment. The transfer function of any peripheral compartment $Z_n(S)$ can be expressed as a fraction of $Z_1(S)$:

$$Z_n(S) = \frac{\kappa_{n,1}}{S + k_n} Z_1(S), \qquad (11)$$

where n = 2, 3, ..., and M.

Thus, for a mammillary model, eqns 10 and 11 are the two generalized transfer functions for the central compartment and all peripheral compartments respectively (Anderson 1984).

Inverse Algorithm

From the principle of degrees of freedom, a set of M-independent functions can uniquely solve for M variables, if the solutions exist. An M-term exponential function consists of 2M parameter values (M coefficients and M eigenvalues). But the eigenvalues must be real, non-zero and unique. Therefore, a set of 2M TRC parameter values can be obtained from the parameter values of an M-term exponential function.

Unlike the mammillary model, all TRC parameter values in a Hepatic model are identifiable. An inverse algorithm for solving a matching set of TRC parameter values from an exponential function was developed. The essential steps of this algorithm are summarized:

(1) Because eqn 9 defines SWBRF as a sum of all CRFs, therefore, $\alpha_{1,1}$ the first exponential term constant coefficient of a CCRF, for example, should be proportional to A_1 the first exponential term constant coefficient of an

M-term exponential SWBRF. Let φ_1 be the proportionality constants of $\alpha_{1,1}$ and A_1 and let $[\varphi]$ be the matrix notation of φ values. Then the relationship for the constant coefficients $\pi_1(t)$ and R(t) can be expressed:

$$\pi_1(t) = [\varphi]\ R(t), \text{ or } \alpha_{n,1} = \varphi_n A_n, \tag{12}$$

again, where n is a numerical index from 1 through M; the number of the exponential terms in the SWBRF. The A_n and $\alpha_{n,1}$ are the n^{th} constant coefficients of SWBRF and CCRF, respectively. In practice, the $[\varphi]$ matrix values consist of a set of M "real" constants. The key for solving this system is to obtain a set of φ values that satisfy the following specifications by an iterative method.

$$-R'(t) = \beta_1\pi_1(t) + \beta_2\pi_2(t) \tag{13}$$

The derivative of a SWBRF is the total excretion from the system. Therefore, eqn 13 is developed from a mass balance law for the Hepatic model because it had two excretion pathways. This equation shows that the negative of the derivative of an SWBRF, which gives the total fraction of an uptake expected to be excreted per day, shall be equal to the sum of excretion rates from two excretion pathways (urine and feces).

(2) Because of eqn 12, the transfer function of the central compartment $Z_1(S)$ can also be yielded via Laplace transformation of a CCRF or an SWBRF as:

$$Z_1(S) = \sum_{n=1}^{M} \frac{\alpha_{n,1}}{S+\gamma_n} = \sum_{n=1}^{M} \frac{\varphi_n A_n}{S+\gamma_n}, \tag{14}$$

The $Z_1(S)$ can be simply expressed as a ratio of two polynomials by multiplying both numerator and denominator of eqn 14 with the lowest common denominator of the equation. That is,

$$Z_1 = \frac{q(S)}{p(S)} \tag{15}$$

If $Z_1(S)$ is obtained from Laplace transformation of an M-term exponential function, the denominator polynomial p(S) should also be an M-order polynomial and the order of numerator q(S) is always one less than the denominator. It is important to assure that q(S) and p(S) have no common factors other than number 1, e.g., H(S) has been reduced to its lowest terms.

(3) Because eqn 10 is a transfer function of a central compartment for solving eqns 1, 2, 3, and 4 analytically using the Laplace transformation method, and because eqn 15 is also presented as a Laplace transformation of an CCRF, numerically these two equations should be equal if they are applied to one system. Thus, by equating the polynomial in the numerator of eqn 10 to the polynomial in the numerator of eqn 15, the following equality is established:

$$\sum_{n=1}^{M} \left\{ \varphi_n A_n \prod_{\substack{i=1 \\ i \neq n}}^{M} (S+\gamma_i) \right\} = \prod_{j=2}^{M} (S+k_j) \tag{16}$$

The right-hand side (RHS) of eqn 16, a simple Π-product of M-1 distinct first-order polynomials associated with k_j, is taken from eqn 10. The left-hand side (LHS) of eqn 16, a combination of "$\sum$" and Π-product

functions, is taken from eqn 15. By setting the RHS of this equation to zero the M-1 roots which exist in the LHS polynomial can be found. Hence, the total removal rate constant k_j's for all peripheral compartments ($j \neq 1$) can be identified.

(4) Similarly, by equating both polynomials in the denominator of eqn 10 and eqn 15, the following equation can be obtained:

$$\left(S+k_1 - \sum_{i=2}^{M} \frac{\rho_i}{S+k_i} \right) \prod_{i=2}^{M} (S+k_i) = \prod_{n=1}^{M} (S+\gamma_n) \qquad (17)$$

Because the k_i's in the LHS of eqn 17 can be solved from eqn 16, the unknowns of this function should be the ρ_i and k_1 only. The RHS of eqn 17 is a Π-product of M first-order polynomials associated with the distinct eigenvalues γ_n. Therefore, the LHS of eqn 17 is a set of M linear equations. These M independent functions can be generated by substituting each root γ_n repeatedly, thereby setting the RHS of the equation to zero. Then, the values of ρ_i and k_1 can be obtained by solving linear functions simultaneously.

It is noteworthy that, in this algorithm, from steps 2 through 4 are similar to that of Landaw et al. (1984). If set $\beta_n=0$ and $n \neq 1$, the above inverse routine can be simplified as the routine described by Bernard (1973). Landaw's is a generalized inverse routine based on a CCRF. Without eqn 12, the routine cannot be useful in our application. Bernard's routine was developed based on a special case of the Hepatic model when $\beta_2=0$. Because $\beta_2=0$, it cannot be used to determine the feces excretion rates via liver-bile secretion pathway.

Urinary and Fecal Excretion Functions

Urinary and fecal elimination processes in the body are really simplified in the proposed model shown in Fig. 3. In addition, because of the first-order kinetic assumption, the amount of material excreted in urine is proportional to its concentration in blood and the amount of material excreted in feces is proportional to its concentration in the liver. Hence, the urinary excretion rate function is simply the CCRF times β_1 and the fecal excretion function is the retention function of the liver compartment times β_2.

However, these simple assumptions must be modified based on physiological observation. Such observation suggests that a large amount of systemic fluid is entering and recycling through the upper part of the small intestine. Therefore the unabsorbed contaminants carried in the fluid which are expected to be excreted from the central compartment will be eventually excreted by the GI tract pathway to feces. Therefore, the modified urinary U(t) and fecal F(t) excretion rate functions then could be defined as:

$$U(t) = (1-\varsigma)\ \beta_1 \pi_1(t), \text{ and} \tag{18}$$

$$F(t) = \varsigma \beta_1 \pi_1(t) + \beta_2 \pi_2(t), \tag{19}$$

where, ς, a branching constant, is the fraction of contents found in the feces that was supposed to be found in urine. In theory, the feces elimination rates should be zero at t=0, then it increases and then decreases in time like

the concentration in Compartment Number 2. Ideally, ζ value can be obtained by solving eqn 19 as:

$$\zeta = - \frac{\beta_2 \pi_2(0)}{\beta_1 \pi_1(0)} , \qquad (20)$$

Since the numerator $\pi_2(t)=0$ at time t=0, this presents a problem for solving ζ using eqn 20. Therefore, perhaps ζ can be obtained from experimental data of a single individual or from a cohort population.

VERIFICATION AND RESULTS

Test Model and Computational Results

For most of the chemical elements, SWBRFs in ICRP-30 (1978) and urinary excretion functions in ICRP-10 (1967) are available for the dose assessment and for the interpretation of intake from urine data, respectively. But there rarely exists a physiological model provided with TRC parameter values which can be used to compute the model's retention or elimination rates simultaneously. Therefore, using the available SWBRFs and/or urine excretion functions to obtain the identifiable TRC parameter values is highly desirable.

To show the properties and demonstrate an application of the proposed model, a four compartment system was established for a later Pu retention-excretion study. The following eight fictitious TRC parameter

values were arbitrary selected by imitating TRC values of Pu in a body for testing purposes. They are;

$$\kappa_{2,1}= 4.0;\quad \kappa_{1,2}= 2.0;\quad \kappa_{3,1}= 5.0;\quad \kappa_{1,3}= 1.0;$$
$$\kappa_{4,1}= 9.0;\quad \kappa_{1,4}= 8.0;\quad \beta_1= 2.0,\ \text{and}\ \beta_2= 0.50. \tag{21}$$

By solving eqns 1 through 8 with the above assigned TRC constants, the following SWBRF was obtained:

$$R(t) = 0.060145\ \text{Exp}(-24.8426361t) + 0.024052\ \text{Exp}(-4.53836203t) + 0.095365\ \text{Exp}(-1.85061157t) + 0.820438\ \text{Exp}(-0.26839173t) \tag{22}$$

where t has a time unit which should be the same as that of TRCs'. Notice that eqn 22 is a four-term exponential function with four distinct real eigenvalues. The sum of the four coefficients in eqn 22 equals one. These were expected properties from solving a four-compartment mammillary model.

The four calculated CRFs' were tabulated in Table 1 by the coefficients with respect to the four eigenvalues, including eqn 22 at the bottom of the table. Suppose eqn 22 is available before eqn 21, then the eqn 22 can be used to identify those TRC values shown in eqn 21 by solving eqns 16 and 17 simultaneously. Therefore, using the eight parameter values in eqn 22 the following TRC parameter values were regenerated. They are:

$\kappa_{1,2}= 2.00009537$; $\kappa_{2,1}= 3.99994493$; $\kappa_{1,3}= 0.99993849$;
$\kappa_{3,1}= 4.99976301$; $\kappa_{1,4}= 8.00008774$; $\kappa_{4,1}= 9.00015831$; and

the excretion rate constants:

$$\beta_1 = 2.00000167 \text{ and } \beta_2 = 0.50001180 \quad (23)$$

All parameter values in eqn 23 are deliberately showed to nine significant digits. In theory, these calculated TRC values in eqn 23 should be identical to those assigned in eqn 21.

Application model and Computation Results:

Leggett et al. (1984) constructed a 12-compartment physiological model to study systemic Pu in the body. They reported a four-term exponential Pu SWBRF for interpretation of bioassay data and assessment of occupational exposure to Pu as follows:

$$R(t) = + 0.012\ \mathrm{Exp}(-0.693t) + 0.02\ \mathrm{Exp}(-0.03t) + 0.042\ \mathrm{Exp}(-0.0028t) + 0.926\ \mathrm{Exp}(-0.0000216t) \quad (24)$$

Because Leggett's SWBRF is a four-term exponential function, it is possible to assume that the SWBRF can be obtained from a four-compartment Hepatic model with a unique set of eight TRC values. The four compartments are one-central compartment for systemic body-fluid and three peripheral compartments for bone, liver, and soft-tissues. Once the TRC parameter values are obtained, the urine and feces excretion rates can be calculated using eqns 18 and 19, respectively. Further, the calculated urine function can be compared with that of Jones (1985) and, likewise the calculated feces function can be compared with that of Langham (1956).

Again, a set of four CRFs were solved using parameter values in eqn 24. These CRF parameter values were tabulated in Table 2 using the same format as that of Table 1. The behavior of the functions was plotted in Fig. 4 from 0.1 to a 1,000-d interval for the following compartment identification purposes.

In Fig. 4, the solid line function starts at the top of the figure, because the uptake in the central compartment is at t=0. Therefore, the solid line describes the retention behavior of the Pu in the central compartment, blood. The three other functions that start at the bottom of figure are the retention predictions for bone, liver and soft-tissues.

Among these three peripheral compartments, the long-dash line function rises and drops within a few days after an uptake, and apparently describes the Pu retention in the soft-tissues. This is true because Pu in the soft-tissues would be expected to have a relatively faster elimination rate in comparison to that in bone and in liver. Furthermore, accepting that Pu in bone has a longer half-time than in liver, the Pu retention in bone is described more appropriately by the short-dash line function, which reflects a rapid initial increase and no sign of decline even after 1,000-d after uptake. Likewise, the double-dotted chain line function which indicates a peak at about 100-d postuptake and a slow decrease would appear to be the best function for describing the Pu retained in liver.

Prediction of Urinary and Fecal Excretion Rates

Leggett reported that 2.4% of the integrated activity in blood may be removed each day in feces. Suppose that the ς parameter in eqns 18 and 19 could be obtained from this suggested value. In this case, ς=0 was used for the calculation of urine and fecal excretion functions. Hence, from Table 2, a set of urinary excretion function U(t) and the fecal excretion function F(t) were calculated:

$$U(t) = + 0.00834388\ \mathrm{Exp}(-0.693t) + 0.00065865\ \mathrm{Exp}(-0.03t) + 0.00003870\ \mathrm{Exp}(-0.0028t) + 0.00001254\ \mathrm{Exp}(-0.0000216t) \quad (25)$$

$$F(t) = - 0.00002811\ \mathrm{Exp}(-0.693t) - 0.00005870\ \mathrm{Exp}(-0.03t) + 0.00007936\ \mathrm{Exp}(-0.0028t) + 0.00000745\ \mathrm{Exp}(-0.0000216t) \quad (26)$$

where the unit of time t is days. Because eqns 25 and 26 were calculated from eqn 24, the eigenvalues in all three equations were expected to be identical.

In 1987, a Pu urinary excretion prediction using the same 12-compartment model as that for eqn 24 was published by Leggett and Echerman (1987). In that paper, Leggett's urine excretion function was not given. But the estimated values were plotted with the Jones' (1984) and a so-called "(Leggett) modified Langham urine function" for the comparison. Because Leggett's Pu urine excretion function is not available, Table 3 is used to tabulate the available data to compare Jones' and eqn 25 estimations.

The estimated urine excretion rates from Table 3 are plotted in Fig. 5: Jones (double dotted-line) and Leggett (dotted-line) and Sun-Leggett, which is eqn 25, (solid-line). Despite the differences of eigenvalues in Jones' function, the plot indicates that the differences among the three models are small: At about 100-d after a single acute uptake, the rates predicted by eqn 25 are well fitted in between by the other two models. From 100 to 10,000-d, the predictions of Sun-Leggett model are slightly lower than Leggett and Jones models, i.e., eqn 25 could provide a moderately more conservative protection factor for interpretation of urine data than the other two models. It is noteworthy that the values predicted from eqn 25 and the Jones model are surprisingly similar from 1,000-d to 10,000-d postuptake.

Usually retention models have to be developed based on the measurements of the organs and tissues, unlike excretion functions which can be obtained simply from the regression analysis of repeated samples. Eqn 24 which describes Leggett's Pu systemic retention function was developed from a multi-compartmental model with a specified set of TRC values interpreted from autopsy results. Jones' urine excretion function was obtained from a statistical analysis of urine data. Fig. 5 shows that Leggett's SWBRF can be transformed through a four-compartment Hepatic model to predict Jones' urinary excretion rate over a 10,000-d period. One could also adjust the ς parameter value to reduce the difference between eqn 25 and Jones or to obtain a desirable urine excretion function based on experimental measurements. This demonstrates that a method using a reliable SWBRF to obtain a reliable urine excretion function is available. This method can be used as a verification technique in the development of retention-excretion models.

According to eqn 19 the Pu excretion rate in feces is the sum of the Pu in the secreted liver-bile and the Pu in a fraction of unabsorbed systemic body fluid. Again, because ς=0 was used, eqn 26 solely described the Pu to be excreted from biliary secretion via GI tract. Eqn 26 has been plotted as the solid-line, and compared with that of Langham as the double dotted-line in Fig. 6. The difference between the two equations is obvious. Langham's fecal excretion function is a monotonically decreasing power function, while eqn 26 is not. Theoretically, however, a function that would yield an increasing and then decreasing excretion would be needed for a description of fecal excretion via the liver pathway.

As mentioned earlier, "a modified Langham urine function" was reported by Leggett and used to compare his Pu urine predictions. The techniques have been applied here for extrapolating the 138-d human data up to 10,000-d. Eqn 26 and Langham's fecal function can be brought together is a k value of 0.04 is used. The Langham fecal function modified in this way is plotted as the dotted-line in Fig. 6. This modified function is not significantly different from the original over the first 100-d period. After that, the modified function departs from the original exponentially as time increases. In Fig. 6, the solid-line indicated as the Sun model is shown as it slowly increases and peaks at about 100-d, then decreases slowly. It crosses Langham's function at about 80-d and merges with Langham's modified function closely from 1,000 to a 10,000-d interval.

It is important to remember that the difference between the two U:F equations plotted in Fig. 1 is directly related to Fig. 5 and Fig. 6. Based upon the Hepatic model's evaluation, Sun's U:F ratio predicted a drastic

decrease followed by an increase, as shown in Fig. 1. It would then be more appropriate for a description of Pu in urine and feces than the description of Langham from the day of uptake to a 10,000-d interval. Due to the scarcity of human data relating to fecal excretion, additional human data will be required to verify the predictions. Because of the over-simplified assumptions that the Pu in the liver, including the liver-bile, is uniformly distributed and that the liver-bile secretion is a continuous process in the body (Steimer et al. 1981), the accuracy of fecal excretion rates obtained from the Hepatic model may be questionable in other applications.

Conclusion

(1) The Hepatic model is a simplified mammillary model which uses first-order kinetics and provides a multi-term exponential solution. The number of the exponential terms in the solution equals the number of compartments constructed in the model. The eigenvalues in the solution are real, non-zero and distinct. The sum of these eigenvalues equals the sum of TRC parameter values in the model.

(2) The Hepatic model has no inverse problem. Because of this unique identifiable property, a set of meaningful TRC parameter values can be obtained from a meaningful SWBRF. If those calculated TRC values can be verified or confirmed by appropriate laboratory animals or human autopsy data, the confidence on the retention or excretion functions could be increased.

(3) Although the Hepatic model is a generalized physiological model, it was constructed for the development of a Pu fecal excretion function. With

some minor modifications, the model may be useful in the development of retention and excretion models of other chemical elements. Further, the Hepatic model can be joined together with the current ICRP-30 GI tract model to develop a new approach for studying the transport phenomena of a contaminant in the enterohepatic circulation. Of course, the new system can improve the prediction on the excretion rates in urine and feces as well.

(4) The Hepatic model can be used to estimate annual and committed absorbed dose in critical organs using a systemic whole-body retention function. The dose rate is calculated by the radioactive transformation rate and the energy absorbed per transformation in an exposed region of interest. The absorbed dose is the sum of energy absorbed per unit mass of a given target tissue for each specific radiation emitted by the radionuclide. Because those organ-specific CRFs computed via an inverse algorithm can be integrated over one year, 50-y, or any interested time interval, the integrated nuclear transformations are proportional to doses that would be received over the integrated time period.

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Table 1. Compartmental retention functions calculated from TRCs of eqn 21 (test model).

	$Exp[-\gamma_1 t]$	$Exp[-\gamma_2 t]$	$Exp[-\gamma_3 t]$	$Exp[-\gamma_4 t]$
γ_i =	24.8426361	4.5383620	1.8506116	0.2683917
	Coefficients of exponential terms			
$\mathfrak{R}_1$:	0.7820848	0.1071431	0.0347404	0.0760317
$\mathfrak{R}_2$:	-0.1400166	-0.2102533	0.2139884	0.1362814
$\mathfrak{R}_3$:	-0.4179134	0.2785640	0.0508447	0.0885049
$\mathfrak{R}_4$:	-0.1640097	-0.1514021	-0.2042083	0.5196200
SUM:	0.0601451	0.0240517	0.0953651	0.8204379

The unit for the rate constant γ is reciprocal time.

Table 2. Compartmental retention functions calculated from parameter values of eqn 24 (Leggett Pu systemic retention function).

	$\text{Exp}[-\gamma_1 t]$	$\text{Exp}[-\gamma_2 t]$	$\text{Exp}[-\gamma_3 t]$	$\text{Exp}[-\gamma_4 t]$
γ_i =	0.693	0.03	0.0028	0.0000216
Organ:	Coefficients of exponential terms			
Blood:	0.9216	0.07275	0.004275	0.001385
Liver:	-0.1035	-0.2159	0.2919	0.02740
Skeleton:	-0.2165	-0.3983	-0.2755	0.8903
Others:	-0.5896	0.5614	0.02148	0.006721
SWBRF:	0.0120	0.0200	0.04217	0.9258

The units for the rate constants γ is day^{-1}.

Table 3. Comparison of plutonium excretion rates in urine sample based on a single injection.

Post-uptake Days	Sun-Leggett[a]	Jones	Leggett	Post-uptake Days	Sun-Leggett[a]	Jones	Leggett
1	4.86E-03	5.10E-03	3.20E-03	110	6.52E-05	8.89E-05	9.00E-05
2	2.76E-03	3.84E-03	1.80E-03	120	5.82E-05	8.02E-05	8.70E-05
3	1.70E-03	3.08E-03	1.20E-03	130	5.27E-05	7.40E-05	8.50E-05
5	8.79E-04	2.31E-03	8.60E-04	140	4.85E-05	6.93E-05	8.20E-05
6	7.31E-04	2.10E-03	6.60E-04	150	4.52E-05	6.56E-05	8.00E-05
7	6.50E-04	1.95E-03	5.30E-04	200	3.62E-05	5.45E-05	7.00E-05
7	6.50E-04	1.95E-03	4.50E-04	300	2.92E-05	4.14E-05	6.00E-05
8	6.01E-04	1.83E-03	3.90E-04	400	2.51E-05	3.27E-05	5.00E-05
9	5.69E-04	1.73E-03	3.40E-04	500	2.19E-05	2.68E-05	4.50E-05
10	5.46E-04	1.65E-03	3.10E-04	600	1.96E-05	2.27E-05	4.00E-05
12	5.12E-04	1.51E-03	2.60E-04	800	1.64E-05	1.80E-05	3.50E-05
14	4.83E-04	1.38E-03	2.30E-04	1000	1.46E-05	1.57E-05	3.00E-05
16	4.57E-04	1.27E-03	2.00E-04	1500	1.27E-05	1.39E-05	2.50E-05
18	4.33E-04	1.17E-03	1.90E-04	2000	1.22E-05	1.35E-05	2.00E-05
20	4.11E-04	1.08E-03	1.70E-04	2500	1.19E-05	1.32E-05	1.70E-05
25	3.60E-04	8.84E-04	1.50E-04	3000	1.18E-05	1.30E-05	1.50E-05
30	3.16E-04	7.25E-04	1.30E-04	4000	1.15E-05	1.27E-05	1.50E-05
35	2.78E-04	5.98E-04	1.25E-04	5000	1.13E-05	1.23E-05	1.50E-05
40	2.46E-04	4.96E-04	1.20E-04	6000	1.10E-05	1.20E-05	1.40E-05
50	1.93E-04	3.47E-04	1.15E-04	8000	1.05E-05	1.13E-05	1.20E-05
60	1.54E-04	2.51E-04	1.10E-04	10000	1.01E-05	1.07E-05	1.10E-05
70	1.25E-04	1.88E-04	1.05E-04	12500	9.57E-06	9.96E-06	1.00E-05
80	1.03E-04	1.47E-04	1.00E-04	15000	9.07E-06	9.27E-06	1.00E-05
90	8.69E-05	1.20E-04	9.70E-05	17500	8.59E-06	8.64E-06	1.00E-05
100	7.46E-05	1.01E-04	9.30E-05	20000	8.14E-06	8.05E-06	1.00E-05

[a] Equation 25 in the text.

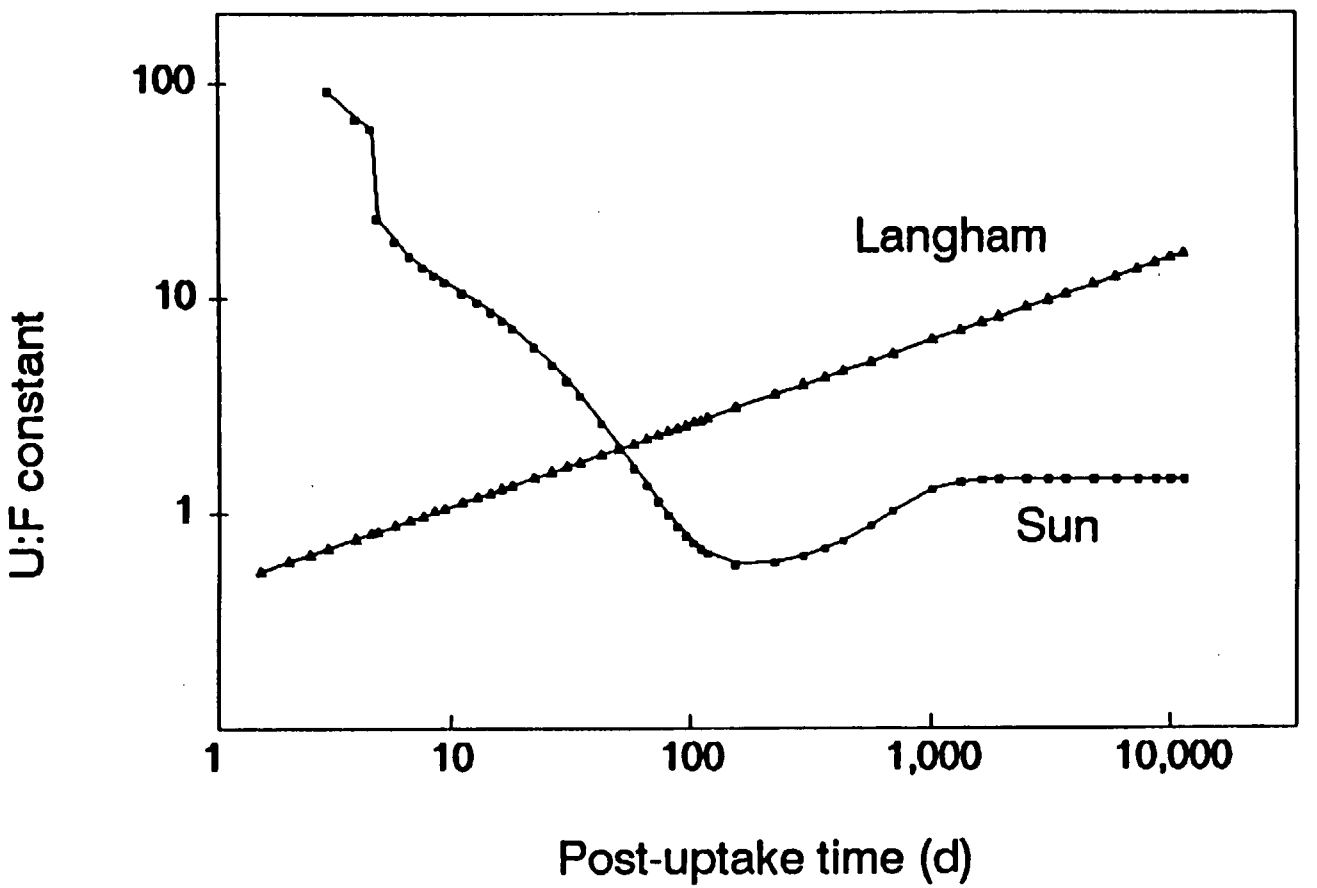

Fig. 1. Predicted ratio of Pu in urine to that in feces from a single injection.

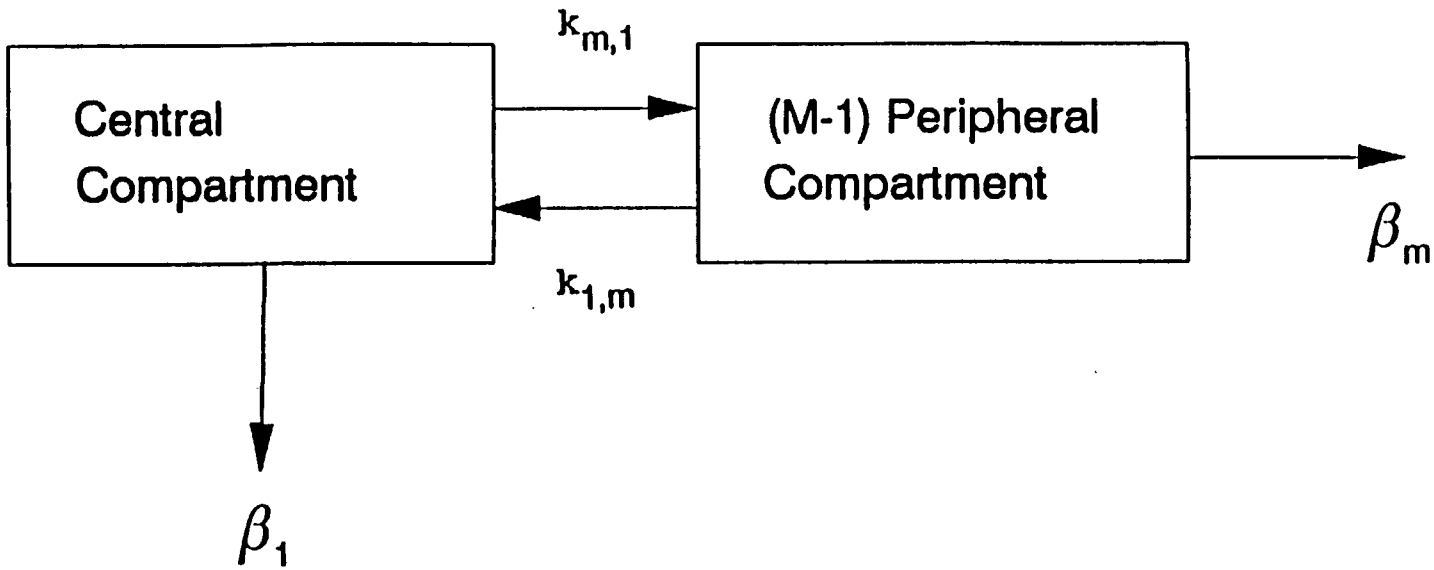

Fig. 2. Diagram of an M-compartment mammillary model and the direction of pathways.

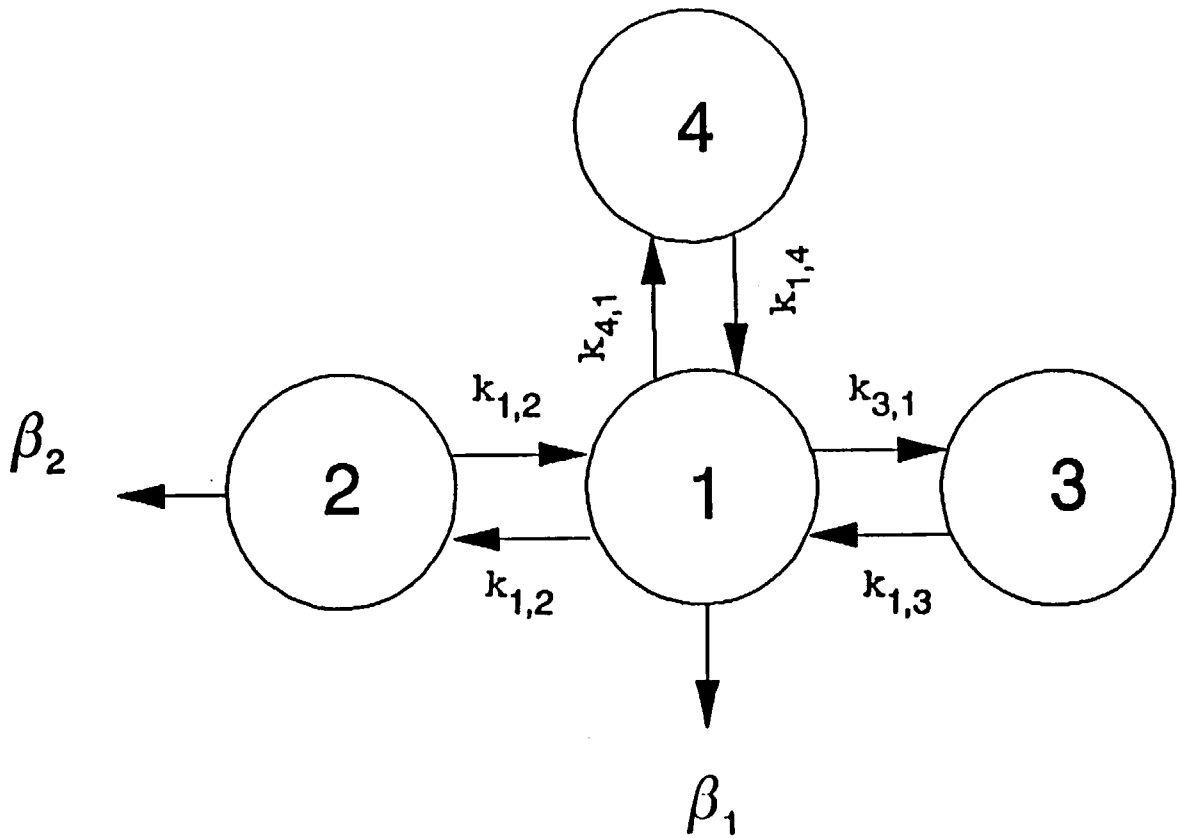

Fig. 3. Diagram of a 4-compartment Hepatic model and the direction of pathways.

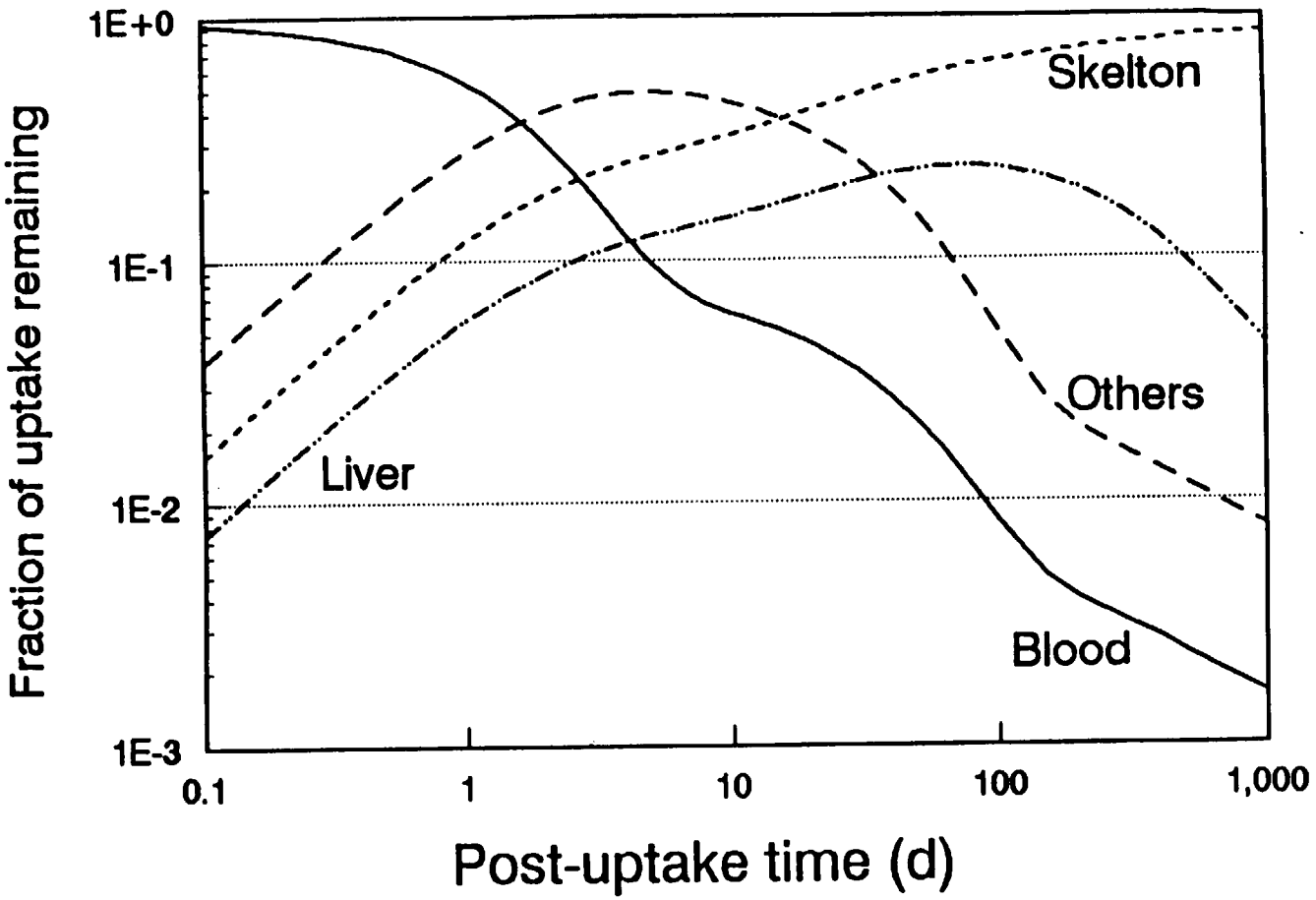

Fig. 4. Comparison of four compartmental retention functions for organs and tissues identification.

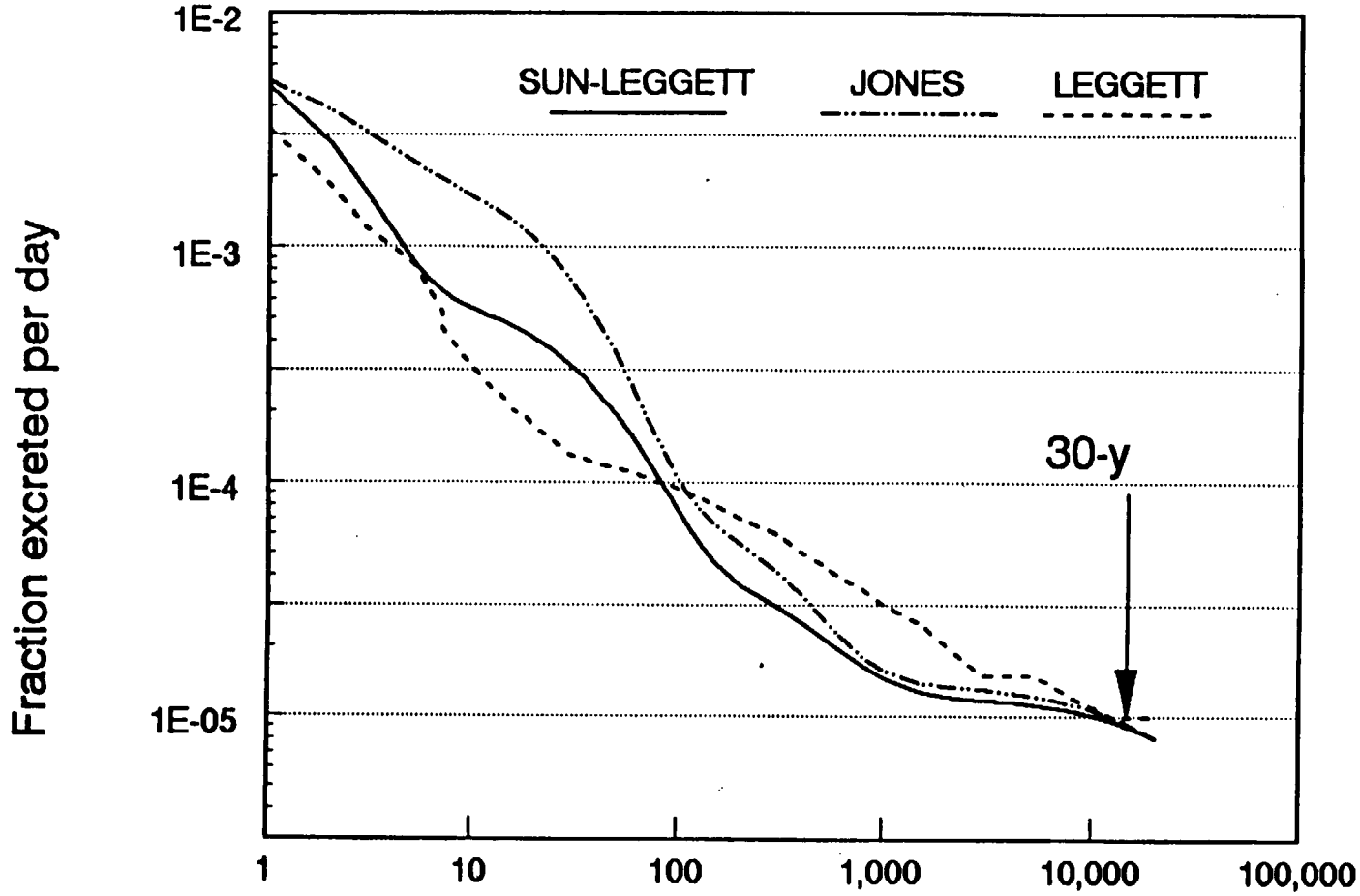

Fig. 5. Comparison of urine excretion functions from a single injection of Pu.

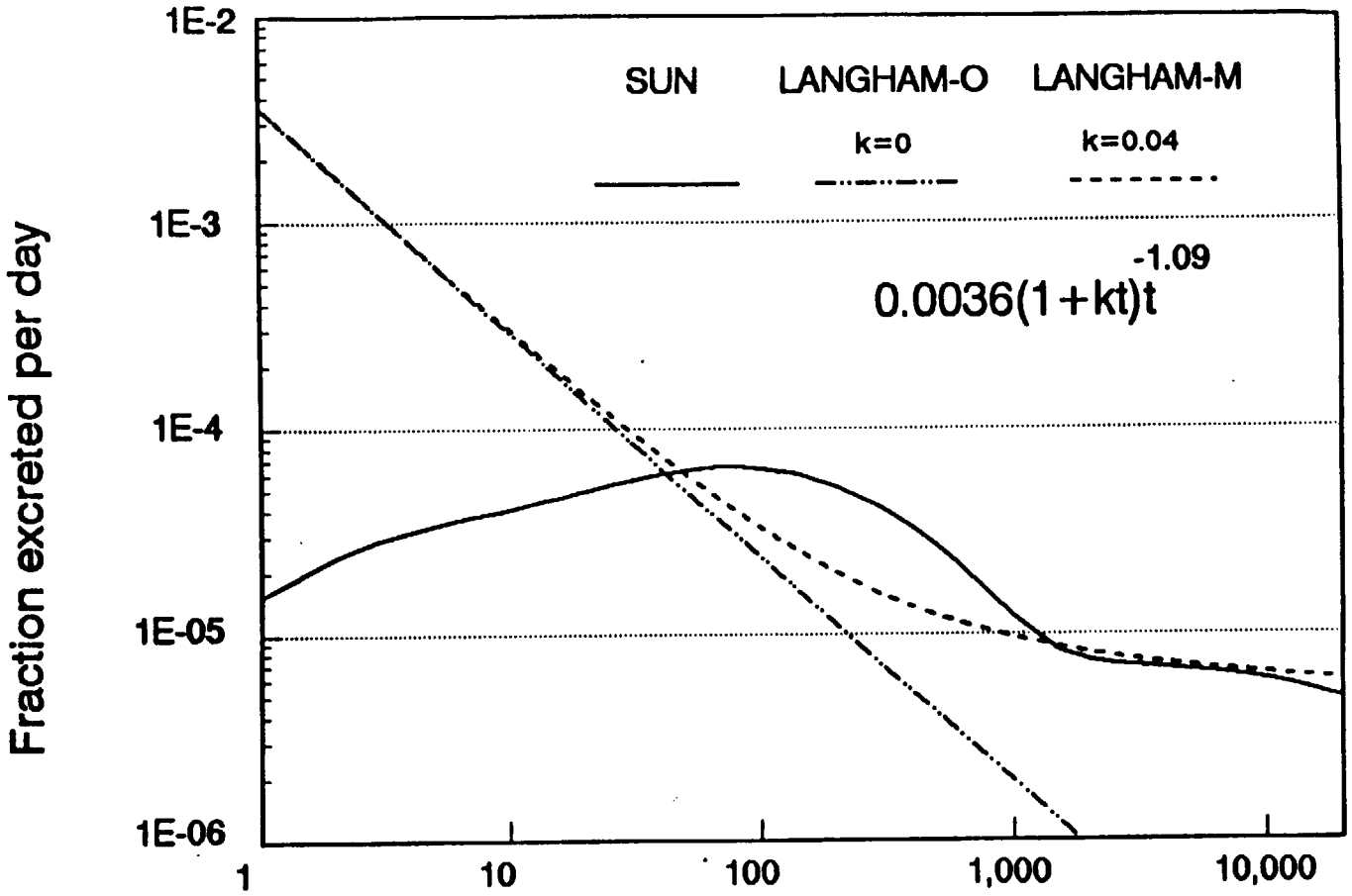

Fig. 6. Comparison of fecal excretion functions from a single injection of Pu.

Bcc: H. Brown

BROOKHAVEN NATIONAL LABORATORY
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Department of Nuclear Energy

Radiological Sciences Division
FAX (516) 282-5810

September 7, 1989

Dr. Patricia Durbin
Lawrence Radiation Laboratory
Building 74B,
University of California
Berkeley, California 94720

Dear Dr. Durbin:

It was a pleasure to meet you and travel with you to the 34th Health Physics Meeting held at the Albuquerque Convention Center. You expressed surprise to hear that your plutonium excretion model is being used for interpretation of urine data and intake estimation for the Marshallese. You pointed out that your model was established in 1972, it is before the time Dr. Rundo completed his long-term plutonium human excretion data. You believe it would be more appropriate interpreting Marshallese urine data using more recent plutonium models. You further expressed that you will write a note to the Health Physics Journal concerning the validity of your plutonium urine excretion model.

The report mentioned above, using your excretion model to interpret the Marshallese urine data for intake, is attached for your information. This report was presented at the Lawrence Livermore National Laboratory by Mr. Bernd Franke, a consultant for the Marshallese.

Again, I enjoyed meeting you and look forward to reading your note in the Health Physics Journal in the near future.

Sincerely yours,

Casper Sun

Casper Sun, PhD

CS:pd
Attachment

BROOKHAVEN NATIONAL LABORATORY
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Department of Nuclear Energy
Radiological Sciences Division

March 14, 1989

Mr. George Taylor
College of Engineering
Texas A&M University
College Station, Texas 77843-3133

Dear George:

As a follow-up of our March 4 telephone conversation on your Rongelap plutonium urine data from the 1988 summer study, I would like to reemphasize that: (1) there are many typos in your data, and (2) you have misinterpreted our urine data for dose calculation.

With regard to ID #206, for example, your list gives a value two order of magnitudes higher than the value in our records. To verify this typo, I have enclosed a copy of your original worksheet for your reference. I would like to thank Mr. Bernd Franke for pointing out this serious mistake. Other errors were found with ID numbers and sampling dates.

Furthermore, you have wrongly treated our 24-hour urine samples as grab samples and increased the estimation of daily Plutonium excretion. It would appear that you have misunderstood our bioassay sampling protocol and our fission tracks calculations. We are concerned that as a result, you have overestimated the intake rates and caused unnecessary concern to the Rongelap people.

Sincerely yours,

Casper Sun

Casper Sun, Ph.D.

CS:pd
Enclosure

cc: H. Brown
B. Franke
H. Kohn
C. Meinhold
J. Poston

Marshall Islands Radiological Safety Program

Whole Body Counting

Brookhaven National Laboratory (BNL) personnel have conducted whole body counting surveys in the Marshall Islands since 1974. These surveys where initiated by a Medical Department team that ran the counting program through 1977. In 1978, counting operations were transferred to the Safety and Environmental Protection Division (SEP) which conducted counting trips to the islands at least once each year through 1985. The 1989 mission was the responsibility of the Radiological Sciences Division.

Through 1979, whole body counting was conducted using a shadow-shielded whole body counter consisting of a stationary crystal and a stationary bed. The crystal was positioned to detect principally radionuclides located in the thorax. In 1980, this counter was replaced by a shadow-shielded chair configuration that detected radioactive material located between the neck and knees. The same crystal and electronics (i.e. power supplies, preamplifiers/amplifiers and ADCs) were utilized with both configurations. For last year's trip, we retained the same crystal-type and shadow-shielded chair configuration, but replaced the electronics as well as the analytical hardware and software.

Whole Body Counting Chair

The chair, which was designed and constructed at Brookhaven, is pictured in Figure 1. It is mounted atop steel plates and is enclosed on both sides and behind by 4" thick, lead-brick walls. The detector is located in a steel cylinder that is mounted on a pivoting arm that is moved across the front of the chair during counting. During counting, the cylinder containing the detector is tilted at an angle of 45° below horizontal toward the person in the chair. The position of the chair can be adjusted both vertically and horizontally.

Three identical chairs were produced. One is in use for personnel monitoring at BNL and the other two are in the Marshall Islands in the whole body counting trailer that is transported on shipboard during counting trips. The chair pictured in Figure 1 is one of the chairs in this trailer. It was intended that calibrations would be done in the BNL chair and made applicable to the field chairs by operating those chairs in the same geometry as had been used during calibration runs at BNL.

Detectors, Electronics and Analytical Hardware and Software

Eleven and one-half inch diameter by four inch thick, thallium-activated, sodium iodide scintillation crystals have been used as detectors since the inception of the program. One of the old crystals and two new crystals, purchased from the same producer as the old one, were used in the field last summer. These crystals are optically coupled to 3-5 inch diameter, low background, high-

gain photomultiplier tubes. The signal outputs are summed and conducted to a Canberra Model 1510 Integrated Signal Processor that contains a power supply, amplifier and analog-to-digital converter (ADC). The ADC, in turn, is connected to an IBM PS/2 Model 70 computer that contains a Canberra S100 multichannel analyzer (MCA) board and Canberra software (S100 and GAMMA-AT) that controls spectral acquisition, display and analysis. This combination of equipment was duplicated at both counting chairs, and a third set of components was brought along for backup in case of equipment failure.

Calibration

As noted above, calibration runs were done using the whole body counting chair at BNL. A BOMAB bottle phantom containing measured amounts of either K-40, or Cs-137 plus Co-60 standard solutions, was used to develop efficiency vs energy calibrations for each of the three detectors that were brought to the Marshall Islands. Activity was uniformly distributed throughout the phantom. Only one of the signal processors and one of the computers were used in the calibration runs. However, prior to the runs, all three signal processors were tested with one of the detectors to determine whether there were significant differences in data acquisition among the processors. None were noted.

The BOMAB bottle phantom consists of head, thorax and pelvic segments along with two arms plus two thigh and two leg segments. By varying the segments used to construct the phantom, three body-size geometries (designated adult, teen amd juvenile) were produced. All nine segments were used to construct the adult geometry. The teen geometry was put together with the head, torso, one thigh and one leg. Two legs, two arms and the head composed the juvenile geometry. The ratio of mass and activity between these geometries is 1 : 0.536 : 0.326 for the adult : teen : juvenile progression. All crystals were calibrated with all three phantom geometries.

In the field, spectra from persons weighing 60 kg or more were analyzed using the adult geometry. Teen geometry was used with spectra from persons weighing 40 gm or more, but less than 60 kg. Spectra from those weighing less than 40 kg were analyzed with the juvenile geometry.

To verify that activity was uniformly distributed in the phantom, an aliquot of the solution from each segment was counted on a lithium-drifted, germanium detector calibrated against an Amersham mixed gamma standard source.

Field Procedures

A document containing the field operating procedures has been appended to this report. See Exhibit_____.

Quality Assurance

During the trip 41 randomly selected individuals were recounted either in the same chair or in the other chair. This represents approximately 4% of the total number of people counted. A separate discussion of these recounts and cross-counts is included in this information packet. Detector efficiencies and resolution were checked each day with Cs-137 and Co-60 check sources. Thirty-six such checks were made at Chair #1 and 34 at Chair #2.

Summary of Results

Last summer's mission was intended to assess the current levels of internal, gamma-emitting radionuclides, particularly Cs-137, in Marshallese populations that had been previously monitored. Stops were made at the islands of Enewetak, Mejato (where the population displaced from Rongelap currently resides), Bikini, Ebeye, Majuro and Utirik. Island residents were invited aboard to be whole body counted. Participation was voluntary. In addition, persons brought on board as participants in the urine collection program were also counted. A total of 977 whole body counts were performed. These included 905 island residents, 5 BNL personnel, 5 crewmen from our support vessel (G. W. Pierce), 5 micronesian nursing students from Pohnpei and Truk who were counted at Majuro, and 13 DOE personnel who either accompanied us on the ship (4) or worked on the islands of Enewetak (2) or Bikini (7). A breakdown of the island residents by sex and age-group (adult, teen, juvenile) is presented in Table 1. In this case, adults are defined as being 16 years of age or older, teens are at least 11, but less than 16, and juveniles are less than 11 years old. The table also shows the number of individuals of each sex and age-group on each island who were found to have Cs-137. The means and standard deviations for the Cs-137 body burdens (nCi), as well as the range and median values for these burdens, in each age-category on each island are shown in Table 2 for males and Table 3 for females.

Table 1. 1989 Marshall Islands Whole Body Counting[a]

	Enewetak	Mejato	Ebeye	Majuro	Utirik
Persons Counted					
Males					
Adults[b]	72	30	46	99	79
Teens[c]	24	12	16	24	19
Juveniles[d]	15	5	4	4	6
Totals	111	47	66	127	104
Females					
Adults	63	26	52	98	71
Teens	34	14	14	27	18
Juveniles	8	4	10	7	4
Totals	105	44	76	132	93
Island Totals	216	91	142	259	197
CS-137 Detected in					
Males					
Adults	68	16	11	58	79
Teens	22	7	1	6	19
Juveniles	13	4	0	0	6
Totals	103	27	12	64	104
Females					
Adults	54	13	7	38	70
Teens	28	2	2	4	18
Juveniles	6	2	4	1	4
Totals	88	17	13	43	92
Island Totals	191	44	25	107	196

[a] Excludes counts of BNL personnel (5), G. W. Pierce crew members (5), DOE personnel accompanying the ship or working on islands (13), and nursing students from Pohnpei and Truk (5) who were counted at Majuro.
[b] Age >= 16 years
[c] Age >= 11 years and < 16 years
[d] Age < 11 years

Table 2. 1989 Marshall Island Mission
Summary of CS-137 Body Burdens (nCi) for Males

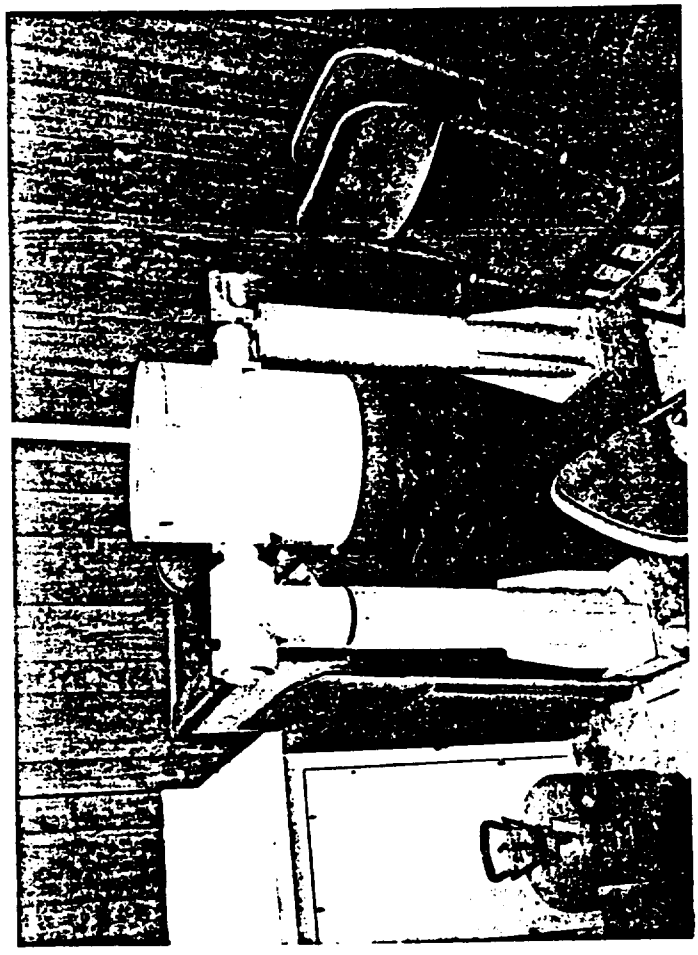
ISLAND		ADULT[A]	TEEN[B]	JUVENILE[C]
ENEWETAK	COUNT	68	22	13
	MEAN	23	16	5.7
	ST DEV	22	22	5.0
	MAX	110	86	19
	MEDIAN	17	8.7	4.7
	MIN	2.6	1.5	1.5
MEJATO	COUNT	16	7	4
	MEAN	3.9	2.3	2.4
	ST DEV	2.3	0.45	0.61
	MAX	12	2.8	3.2
	MEDIAN	3.5	2.3	2.3
	MIN	1.9	1.5	1.8
EBEYE	COUNT	11	1	0
	MEAN	4.8	1.6	
	ST DEV	3.6		
	MAX	13	1.6	
	MEDIAN	3.1	1.6	
	MIN	1.9	1.6	
MAJURO	COUNT	58	6	0
	MEAN	16	10.0	
	ST DEV	16	1.7	
	MAX	90	45	
	MEDIAN	6.9	2.6	
	MIN	1.7	1.6	
UTIRIK	COUNT	79	19	6
	MEAN	40	44	31
	ST DEV	21	20	11
	MAX	95	80	52
	MEDIAN	34	40	26
	MIN	5.2	1.6	23

[A] Age >= 16 yrs
[b] Age >= 11 yrs and < 16 yrs
[c] Age < 11 yrs

Table 3. 1989 Marshall Islands Mission
Summary of Cs-137 Body Burdens (nCi) for Females

ISLAND		ADULT[a]	TEEN[b]	JUVENILE[c]
ENEWETAK	COUNT	54	28	6
	MEAN	11	9.1	5.0
	ST DEV	7.6	7.5	2.9
	MAX	43	39	7.9
	MEDIAN	9.6	6.5	5.2
	MIN	2.0	1.3	1.5
MEJATO	COUNT	13	2	2
	MEAN	3.7	3.1	2.4
	ST DEV	1.5	0.6	1.6
	MAX	5.8	3.5	3.5
	MEDIAN	3.6	3.1	2.4
	MIN	2.0	2.7	1.2
EBEYE	COUNT	7	2	4
	MEAN	4.8	1.7	1.9
	ST DEV	7.3	0.16	0.4
	MAX	21	1.8	2.2
	MEDIAN	2.0	1.7	2.1
	MIN	1.5	1.6	1.3
MAJURO	COUNT	38	4	1
	MEAN	10.0	6.2	2.5
	ST DEV	11	5.4	
	MAX	42	14	2.5
	MEDIAN	5.0	4.7	2.5
	MIN	2.0	1.4	2.5
UTIRIK	COUNT	70	18	4
	MEAN	29	26	29.1
	ST DEV	15	14	6.5
	MAX	70	62	36
	MEDIAN	27	25	29
	MIN	3.7	4.8	23

[a] Age >= 16 yrs
[b] Age >=11 yrs and < 16 yrs
[c] Age < 11 yrs



WHOLE BODY COUNTING INSTRUCTIONS

INTRODUCTION

Before we begin a detailed description of the daily procedure for using whole body counting (WBC) hardware/software let us take a brief overview of the entire WBC process. WBC essentially consists of several simple steps.

First the participant is allowed to seat her/himself comfortably into the WBC chair (a good sense of humor is recommended).

Next you use your software, mostly in the MS/WINDOWS environment. But before you get into WINDOWS you first use a pop-up program to tell each computer it is connected to a LaserJet printer through the 5-th Generation Logical Connection (this is the little box with red wirings running to the computers). Then you enter the S100 program, telling it some basic information and starting it on its way actually controlling and performing the counting process.

While S100 chugs along (for about 10-15 minutes) you invoke a special WINDOWS program which produces a cardfile system into which you enter some anecdotal information about the participant currently in the WBC chair.

When S100 finishes its work it flashes a message to you saying all has gone well and it is ready for the next participant -- but wait -- you must first pass this information collected by S100 to another piece of software called GMAT, which analyses the information and produces a computer output which you give to the participant.

Lastly you thank the Marshallese guest, who leaves the counting laboratory. You are then ready to repeat the process with the next participant.

THESE CAVEATS AND RULES SHOULD BE FOLLOWED SCRUPULOUSLY!

1. Each participant will be weighed and height measured prior to the beginning of the counting procedure.

2. All Canberra hardware/software settings can ONLY be changed by either Jim, Leo, or Casper. A LOG OF ANY SUCH CHANGES MUST BE MAINTAINED, including previous and new settings, as well as date, time, and reasons(s) for any changes.

3. DURING the counting process you will have approximately 15 minutes to complete a "Cardfile" entry on the participant being measured and to update the GAM do-loop. This card entry is a VITAL part of the data. Each person's card must have a title which corresponds EXACTLY to the title given to the file containing that person's counting data.

   Also note that you MUST complete each card's entries BEFORE the counting finishes. Otherwise the S100 software will interfere with the card entry process: a message will flash on the screen that counting has been completed, and you will find that much of your card entry information has not been saved. If this happens you will have to complete the card entries BEFORE going on to the next participant!

4. If an unexpected message appears on the screen while working in a menu or other program such as "ACARD," the situation can be cleared by using the SPACEBAR which will return you to your working menu and possibly one or more "ALT + ESC"s.

Note: in what follows the notation "ALT + ESC" means simultaneously hold down the Alt and ESC keys."F10" means type the Function 10 key.

FORGET about the quotation marks!

POWERING UP THE CANBERRA SYSTEM AND DOING WBC

1. System Power-up begins in the DOS root directory. The first step is to configure the 5-th Generation Logical Connection, telling it that the computer is connected to the LaserJet (or the dot matrix printer, whichever you prefer). This is done by using a macro as follows:

   Type "FIFTH" "ENTER"

   Examine the menu box that is displayed to see that the arrow in the second column is pointing to HP Laserjet. If it is not use the up/down arrow keys to move the highlight band to the HP Laserjet setting. Then type "ENTER". The display will show:

   c:\ > print _____.prn
   Name of list device [PRN]

   Type LPT1 "ENTER"

   Type PTEST "ENTER"

   The laser printer should do a form feed.

2. You now start S100: type "START". This batch file is designed to perform all the necessary steps to display the "Data Aquisition Menu" on the computer screen. The "MCA Menu" will display the following message: "Welcome to the Canberra System 100" and display a memory assignment window. In the lower left corner of the screen will be an icon for the "DOS Executive."

   Check that the "Assignment Window" directory indicates that "ADC#1" and "4096 Full Memory Size" are selected. CHECK WITH JIM, LEO, OR CASPER IF ANY OF THESE SETTINGS ARE NOT CORRECT. DO NOT PROCEED UNTIL YOU RESOLVE THIS.

2. Next clear the "Assignment Window" by **depressing the SPACEBAR once.**

3. Type "Alt + L" and then "V" to display the following "view" window which describes the order of the WBC process:

VIEW
001 Clear, Group: First 8th, Data 002 Preset, Group: First 8th, Live Time: 900 sec 003 Aquire, Group: First 8th, Start 004 Save, Group: First 8th, Data: Spectrum, Device: Disk, SDnnnn.MCA 005 End of Task
Current Cycle: 1 Preset Cycles: 1 [OK]

NOTE: THIS WINDOW SHOULD BE PRESET AS ABOVE ... PLEASE CHECK THAT THIS IS INDEED THE CASE, AND NOTIFY JIM, LEO, OR CASPER IF IT IS NOT. DO NOT PROCEED UNTIL YOU RESOLVE THIS.

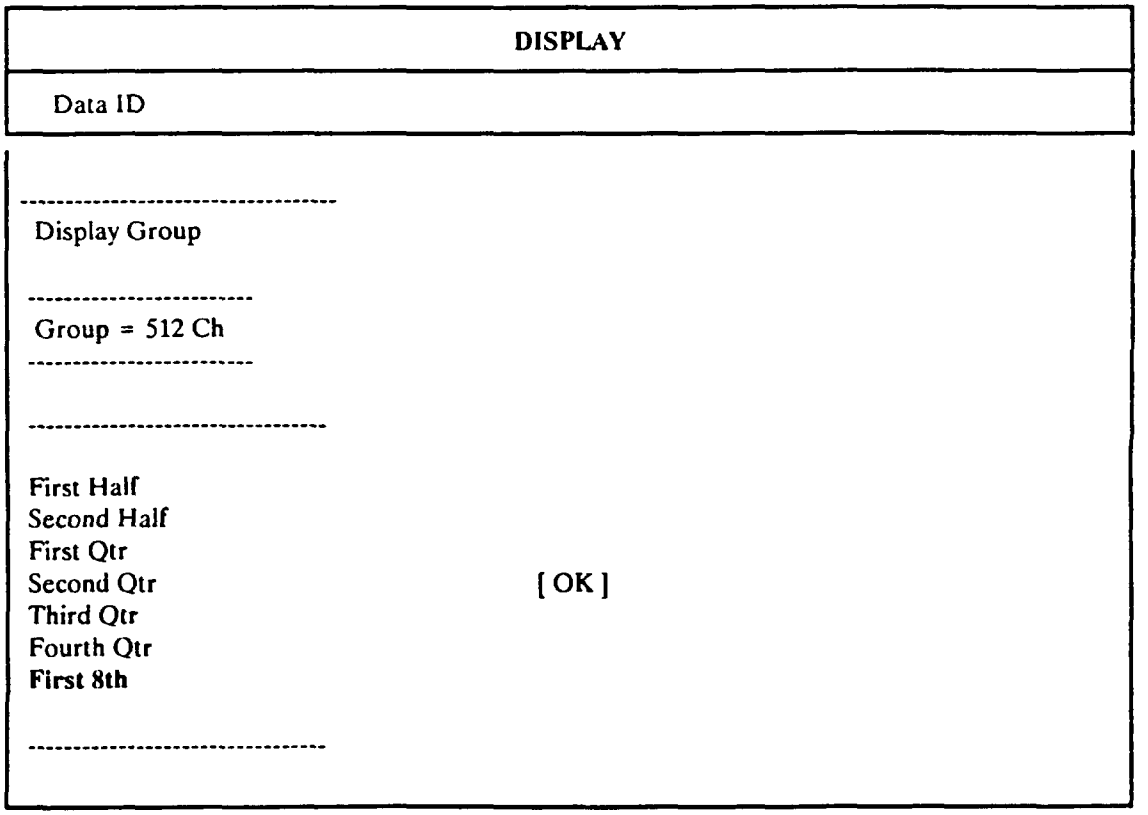
IMPORTANT FOR FIRST TIME USERS: please check with Jim, Leo, or Casper, to understand the meaning and use of these settings!

3.1 If the settings in the "view" window are correct then type **SPACE** to proceed with Task 4. below ("Executing the WBC Task"). Otherwise go to step 3.2 below.

3.2 Type **SPACE**, "**ALT+L**", "**N**", "**F10**", "**F**", "**O**", (use TAB and arrow keys to highlight) "**MARSH8TH.TSK**", "**TAB**", "**TAB**", **SPACE, SPACE.**

4. Executing the WBC Task

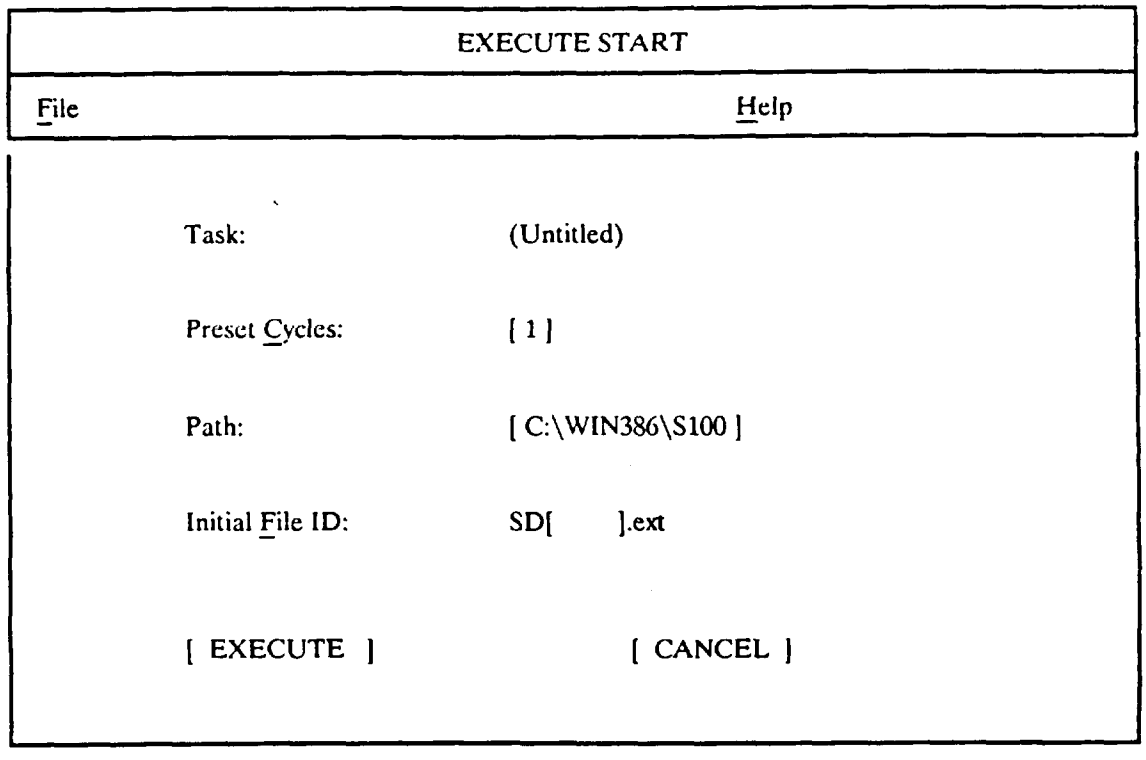
4.1 Type "**ALT+D**", "**G**" to verify that the correct Display Group (i.e., "First 8th") has been selected to work with. You will invoke a window looking similar to this:



4.2 Set the group selection to the highlighted group (i.e., "First 8th") if necessary.

> **NOTE:** Initially the cursor is in the "OK" box. To get to the "First 8th" position you must use the **"TAB"** to place the cursor in the "Display Group" selection box. Once there you use the arrow keys to move the cursor to the desired group (i.e., "First 8th") , followed by the **SPACEBAR** to set the selection. Then **"TAB"** to the "OK" box and type **"RETURN"**.

4.3 Type **"Alt+L", "S"** to display the window entitled "Execute Start" menu. It should look something like this:



> **NOTE: make sure the participant is seated and all equipment is ready for acquisition before proceding with this step!**

Before you begin execution of this task (by typing the **SPACEBAR**) you must ascertain that the information in the window is correct. That is:

4.3.1 **TAB**, **TAB** (which should highlight "Preset Cycles") and enter "1".

4.3.2 **TAB** (to highlight "Path"): it should be set up to "C:\WIN386\S100".

4.3.3 **TAB** highlights the Initial File ID. Enter the appropriate filename. This should correspond to the entry on the index line in the cardfile [see the ACARD section]. PLEASE CHECK WITH JIM, LEO, OR CASPER ABOUT THIS!

File names will be 4-digit numbers of the form XNNN where X identifies the chair (for the left chair use odd numbers starting with one, and for the right chair use even numbers starting with two), and where NNN is a 3-digit number starting at 001. Once this 4-digit number is entered the software will automatically update it after each count, unless the computer is reset.

4.3.4 **TAB**, SPACE begins execution of the WBC process. **IF YOU ARE UNSURE FOR ANY REASON SIMPLY TAB OVER TO THE "CANCEL" BOX AND TYPE SPACE.**

5. Information entries while WBC is working:

5.1 **Spectrum Header:** Access **Spectrum Header Entry** by typing "Alt + D", "G", "F10", "D", "D".

- Enter the following information separated by blanks: the person's ID#, their last name, first name, island, sex, and age.

  EXAMPLE:3124 THOMSON,PAUL T M 34

  (Note that there is no space after the comma)

  Use the following ISLAND index codes:

  T - Mejato (Rongelap)
  U - Utirik
  E - Enewetak
  M - Majuro
  B - EBeye

- Exit to the main MENU with the following sequence: "Tab", "SPACE", "SPACE". This completes the information header for the file which will contain the whole body measurements.

5.2 CARDFILE entries while WBC is working:

- Remember, each CARDFILE information sheet corresponds to a participants' WBC datafile. Specifically, we are now going to spend the next few minutes entering information about the participant who has just started the WBC process.

- Each person will have an index card with name, island, sex, age, weight, height, urine group (Y or N), and background information.

- To select **CARDFILE** (which is run from file "ACARD" in DOS) enter the following sequence: "**Alt+ESC**", "**Alt+SPACEBAR**", "**R**". An alternative after the first usage of this program is to type two "**Alt+ESC**"s followed by an "**Alt+F5**". This opens the DOS Executive.

- Type "A" until "ACARD" is highlighted and then type "**ENTER**". You will see an image of a deck of index cards (with the most previously completed at card at the front of the deck).

- Type "**F7**" to add a new blank card. This opens the index line. Enter the same information as was recorded in "Spectrum Header" in Task 5.1. Press "**RETURN**". Now type the information for the body of the card (i.e., date of birth, age, weight, height, urine program (Y or N), and background information). Fill in required data using the cursor to align your information. Type "**F6**" to reopen the Index Line. Type "**RETURN**" each time you want to go to the next line.

  Note that if you continuously enter data a carriage return will automatically be inserted for you by the CARDFILE program. Use the **control arrows** to position the cursor anywhere on the display screen.

  The **BACKSPACE** key deletes characters behind the cursor. When the **NUMBER LOCK** key is OFF then the key pad **DELETE** key will also delete characters under the cursor.

- The information on the index line should be identical to what you typed in Task 5.1 ("Spectrum Header").

- To save what you have written BEFORE EXITING type "**ALT+F**", "**S**".

- To exit CARDFILE so you can access another program type "**ALT+F**", "**X**", "**S**". You will be asked to confirm that you want to save what you have just typed.

- Use "**ALT+F9**" to convert ACARD to an icon between uses.

- Make the DOS Executive an icon by typing "**ALT+F9**" while the DOS Executive window is open.

- To return to the Canberra S100 Data Collection Screen type "**ALT+ESC**".

SPECTRAL ANALYSIS

6. Reports of spectra obtained during the day are generated at the end of the day by using program GAM. Use program GAM for automated analyses of S100 generated data. Remember that what results from this process is a written report which will be given to the participant.

For all analyses, the individuals counted will be classified into categories based upon their body weight, as follows:

Category	Weight Limits
Adult	≥ 60 kgm
Teenager	≥ 40 kgm and < 60 kgm
Juvenile	< 40 kgm.

The geometry (i.e., efficiency) files and environmental background files used in analyses are specific to individual categories. Therefore, spectra from individuals of a particular category must be analyzed using geometry and background files appropriate to that category, and separate analyses will be required for "adult," "teenagers," and "juveniles." **It is therefore important to ascertain that the appropriate geometry and environmental background files are being used before starting analyses for a given individual.**

6.1 GAMMA-AT, the program used for spectral analyses, is run from DOS rather than WINDOWS. Assuming that you are starting from a condition with the S100 window open, and ACARD and the DOS Executive displayed as icons, GAMMA-AT is set up as follows:

1. Close S100 by typing "**ALT+F4**"

2. Open the ACARD window by typing "**ALT+ESC**". Save ACARD by typing "**F10**", "**F**", "**S**". Then close ACARD with an "**ALT+F4**".

3. When ACARD is closed the DOS Executive window will be open. Type "**ALT+F4**" which reveals an information box informing you that "This will end your windows session." Type "**RETURN**" to close the window and reenter DOS.

4. The DOS prompt will read C:\WIN386\S100>

   a. Change directories by typing "**CD\GMAT**","**ENTER**".

   B. When the new prompt C:\GMAT> appears, start GAMMA-AT by typing "**GAM**", "**RETURN**".

6.2 This brings up a list of analysis parameters as follows:

Use MARSHALL library.
MCA number 0.
Subtract environmental activity file using file number 3102.
Search for unidentified peaks.
Apply gain-shift compensation.
Report LLD values with peak confidence 95.00% (1.645 sigma).
512 channels in the spectrum.
Error quotation = 1.00 sigma.
Maximum percent uncertainty in activity = 100%.
Hard copy output to SC.

Use these parameters? (Y,N): ___

Type **"Y", "RETURN"** if all parameters listed are correct for the analysis to be completed, and proceed to the discussion of the DO function below (section 6.3).

In general, the only parameter that needs to be changed at this time is the environmental activity file number. These files are specific to both the island and the geometry, so be careful that you use the correct file number. If the number must be changed type "N", **"RETURN"** and proceed with the instructions immediately below.

Query	Response
MCA number? [0]:	**"RETURN"**
Analysis library? [MARSHALL]:	**"RETURN"**
Subtract environmental activity? (Y,N):	**"Y", "RETURN"**
Environmental activity file number? [3102]	**"xxxx", "RETURN"**
Search for unidentified peaks? (Y,N):	**"Y", "RETURN"**
Apply gain-shift compensations.	No response needed
Report LLD values? (Y,N):	**"Y", "RETURN"**
Peak confidence level? [95.0%]:	**"RETURN"**
Error signal? [1.00]	**"RETURN"**
Maximum percent uncertainty in activity? [100]:	**"RETURN"**
Hard copy output device (Scree,Printer,None):[S]"	**"P"**
Output reports to disk? (Y,N) [N]:	**"RETURN"**
Save these values in parameter file? (Y,N):	

If all entries are correct, type **"Y", "RETURN"** and proceed to the DO function below (section 6.3). If, however, incorrect entries have been made, type **"N", "RETURN"** and, when the GAM> prompt appears, type **"IN", "RETURN"**. This will enable you to restart the dialogue shown above after you respond to the following questions.

Query	Response
Read default parameter file? (Y,N):	**"Y","RETURN"**
Show current parameters? (Y,N):	**"Y","RETURN"**

The original list of analysis parameters will be displayed. Respond **"N","RETURN"** to the "Use these parameters? (Y,N)?" query, and then redo the dialogue shown above.

6.3 DO Function: You will now get a larger "GAM" window with the following prompts, to which you will answer as indicated in the "RESPONSE" column below:

PROMPT:	RESPONSE: (followed by a "RETURN" unless indicated otherwise)
GAM>	**DO**
System 100 files? (Y,N):	**Y**
Enter S100 path [\WINDOWS\S100]	**WIN386\S100**
First Spectrum Number:	**Starting SDnnnn.MCA file #**
Final Spectrum Number:	**Ending SDnnn.MCA file #**

(up to 50 consecutively numbered files can be analyzed)

Hard copy output device <Screen,Printer,None>[P]: **RETURN**

[P] is the default response selected by a carriage return. Select a new option by entering its first character.

Output reports to disk? <Y,N> [N]: **RETURN**

Detector number? [X]: **X = Detector ID number**

New selection becomes default number accessed with a carriage return.
(398 at chair #1, 400 at chair #2)

Geometry number? [Y]: **Y = Geometry ID number**

New selection becomes default number accessed with a carriage return
(Chair #1: "Adult" = 6; "Teen" = 7, "Juvenile" = 8)
(Chair #2: "Adult" = 2; "Teen" = 3, "Juvenile" = 4)

Use Calibration from Spectrum Data Files? <Y,N> **N**

(Use calibration from detector/geometry files instead.)

Analyzed by: _____ **Enter operators initials**

For S100 file SDxxxx.MCA enter the decay correction interval (M 0.000000E-01): **0**

Repeat for every spectrum to be analyzed and printed.

Wait for files? (Y,N): **N**

Analysis now begins. As each file is analyzed, the report will be printed and the following display appears:

Analysis of S100 data from file C:\WIN386\S100\SDxxxx.MCA
Analyzing

After all analyses and reports are finished, the following dialogue appears:

Hard copy output device (Screen,Printer,None)?[S]:	**"P","RETURN"**
Output reports to disk? (Y,N)[N]:	**"RETURN"**

The procedure is now complete and the prompt GAM> appears.

To restart the procedure for another geometry setting, type "IN","RETURN" and repeat the process starting at 6.2 above.

7. Putting the system to bed for the night [OR IN CASE OF A PLANNED SHUTDOWN].

7.1 Starting in the "CANBERRA MENU" type "**ALT+SPACE**", "**C**".

[**For the first time only**, go to the "GMAT MENU" and enter "**Y**", "**RETURN**".]

7.2 Next type in "**EX**" or "**ex**", and "**RETURN**".

7.3 You should default to the "DOS" window where you need to enter "**ALT+F4**", **SPACE**.

8. Daily System Backup. This task will be performed only by Jim, Leo, or Casper.

At the end of each day before powering down it is necessary to backup all WBC data files and their corresponding ACARD files onto the D: hardisks and diskettes. One week's data from each chair fits on a single high density diskette. So we will be amassing a collection of diskettes, one for each week's data collection from each chair, and a collection of files in two subdirectories on the D: hardisks. The backup procedure will be done in DOS without WINDOWS. Do the following daily for each computer:

8.1 On the first day of each week insert a new, formatted diskette into the A:-drive. On each subsequent day of the week reinsert that week's backup diskette into the A:-drive.

8.2 Type "**HOME**" to take you to root directory.

8.3 Type "**BAKUP**" (a macro) to perform the actual backup procedures. Messages will appear to indicate that the backup procedures have indeed worked.

8.4 Remove the diskette from the A:-drive and store it in a safe place.

8.5 Shut off the system.

NORMAL UP-AND-RUNNING OPERATING SEQUENCE (these instructions take effect starting with the second count)

1. Type "Alt+L", "S" to display the window entitled "Execute Start" menu.

NOTE: make sure the participant is seated and all equipment is ready for acquisition before proceding with this step!

2. SPACE begins execution of the WBC process.

3. **Information entries while WBC is working:**

3.1 **Spectrum Header:** Access **Spectrum Header Entry** by typing "Alt+D", "G", "F10", "D", "D".

- Enter the following information separated by blanks: the person's ID#, their island, sex, age, and name.
- Enter the following information separated by blanks: the person's ID#, their last name, first name, island, sex, and age.

  EXAMPLE:3124 THOMSON,PAUL T M 34

  (Note that there is no space after the comma)

  Use the following ISLAND index codes:

  T - Mejato (Rongelap)
  U - Utirik
  E - Enewetak
  M - Majuro
  B - EBeye

- Exit to the main MENU with the following sequence: **"Tab", "SPACE", "SPACE".** This completes the information header for the file which will contain the whole body measurements.

3.2 **CARDFILE entries while WBC is working:**

- Remember, each CARDFILE information sheet corresponds to a participants' WBC datafile. Specifically, we are now going to spend the next few minutes entering information about the participant who has just started the WBC process.

- To select **CARDFILE** (which is run from file "ACARD" in DOS) enter the following sequence: "**Alt+ESC**", "**Alt+ESC**", "**Alt+SPACEBAR**", "**R**". An alternative after the first usage of this program is to type two "**Alt+ESC**"s followed by an "**Alt+F5**".

- Type "**A**" until "ACARD" is highlighted and then type "**RETURN**". You will see an image of a deck of index cards (with the most previously completed at card at the front of the deck).

- Type "**F7**" to add a new blank card. Fill in required data using the cursor to align your information. Type "**F6**" to open the Index Line (if you are not already there) or to reopen the Index Line. Type "**RETURN**" each time you want to go to the next line.

  Note that if you continuously enter data a carriage return will automatically be inserted for you by the CARDFILE program. Use the **control arrows** to position the cursor anywhere on the display screen.

  The **BACKSPACE** key deletes characters behind the cursor. When the **NUMBER LOCK** key is OFF then the key pad **DELETE** key will also delete characters under the cursor.

- The information on the index line should be identical to what you typed in Task 5.1 ("Spectrum Header").

- When you're through type "**ALT+F**", "**X**" to exit "ACARD".

- **To save what you have written** BEFORE EXITING type "**ALT+F**", "**S**".

- To exit CARDFILE so you can access another program type "**ALT+F**", "**X**", "**S**". You will be asked to confirm that you want to save what you have just typed.

- To return to the Canberra S100 Data Collection Screen type "**ALT+ESC**".

4. To use program GAM for automated analyses of S100 generated data. Remember that what results from this process is a written report which will be given to the participant.

4.1 Select GAM by typing "**ALT+ESC**", "**ALT+SPACE**", "**R**".

4.2 Now you will get a "GAM" window with the following prompts, to which you will answer as indicated in the "RESPONSE" column below:

PROMPT:	RESPONSE: (followed by a "**RETURN**" unless indicated otherwise)
GAM>	**DO**
System 100 files? (Y,N):	**Y**
Enter S100 path [\WINDOWS\S100]	**WIN386\S100**
First Spectrum Number:	**Starting SDnnnn.MCA file #**
Final Spectrum Number:	**Ending SDnnn.MCA file #**
Hard copy output device <Screen,Printer,None>[P]:	**RETURN**

[P] is the default response selected by a carriage return. Select a new option by entering its first character.

Output reports to disk? <Y,N> [N]:	**RETURN**
Detector number? [X]:	**X = Detector ID number**

New selection becomes default number accessed with a carriage return. (Chair #1 is 398; Chair #2 is 400)

Geometry number? [Y]:	**Y = Geometry ID number**

New selection becomes default number accessed with a carriage return. Select the proper geometry file according to the body weight of the individual, as follows:

Counted Geometry	Weight	Geometry # Chair #1	Chair #2
Adult	> 60 kgm	6	2
Teen	>40 & <60 kgm	7	3
Juvenile	<40 kgm	8	4

Use Calibration from Spectrum Data Files? <Y,N>	**N**
Analyzed by: _____	**Enter operators initials**
For S100 file SDxxxx.MCA enter the decay correction interval (M 0.000000E-01):	**0**

Repeat for every spectrum to be analyzed and printed.

4.3 When the analysis is printed out exit to the S100 menu by typing "**ALT-ESC**" to start the process with the next participant.

The following steps are for information purposes only, and should be used only if you get a message like "ACARD DOESN'T EXIST"

1. To access the S100 directory type "**ALT+ESC**", "**ALT+ESC**", "**ALT+SPACE**", "**R**". An alternative procedure is to type two "**ALT+ESC**"s followed by "**F5**".

2. To close a window to change the directory type "**ALT+S**", "**C**"

   To create ACARD it is necessary for the path to be: **C:\WIN386\S100**. This is done by highlighting the path instruction and typing the following:

   Type five (5) "**BACKSPACES**", and "**RETURN**" to put you in the \WIN386 directory.

3. To execute cardfile type "**C**"s until CARDFILE.EXE is highlighted, then type "**RETURN**".

4. Type "**ALT**", "**F**", "**A**" and then tab over to the Open Save File Name which you want to say:

   \WIN386\S100\ACARD

   ---

   to save ACARD.CRD on DIR \win386\s100

5. To exit CARDFILE type "**ALT**", "**F**", "**X**".

6. Type "**UP-ARROWS**" to highlight S100, then type "**RETURN**" to put back in the S100 Directory

7. Type "**DOWN-ARROW**", "**ALT+ESC**" to return us to the S100 Data Collection screen.

Exploratory Data Analyses

NOTE

In the following text results have been calculated using original and censored data [that is, Cs-137 whole body counts (WBC) which were above MDL.

1. General: Table 1A summarizes data for each island where results are shown irrespective of which chair was used for WBC. Table 1B is a similiar summary where we have broken out each island's data in terms of particular WBC chairs.

2. Question: Are the distribution of [censored] counts normal?

For data from most islands (i.e., Enewetak, Medrin, Majatto, Ebeye) the distribution of counts is highly skewed and more peaked (i.e., kurtosis is high) than would be expected if the distribution were normal. As a standard procedure the distribution of counts are often logged and then compared to see if a lognormal distribution results. This happened as expected, with results as shown in Table 2.

3. Question: (H_o) Are measurements [censored] taken in Chair 1 different from measurements [censored] taken in Chair 2?? (I.e., are measurements in Chairs 1 and 2 from same population distribution?)

A 2-tailed t-test was performed for data from both chairs taken at each island (using raw and transformed data). For both types of data (i.e., raw and log-transformed), at the 99% level of confidence there was no significant difference between the average or variance of measurements taken between chairs; that is, it appears as though the average and variance of WBC measurements from Chair 1 have the same distribution as average and variance measurements from Chair 2. Figures 1a-e illustrate the distribution of Cs-137 activities measured in each chair at each island. Figure 2 illustrates the values of the mean and median, and the 10th- and 90th-percentiles for each chair at each island.

4. Question: (H_o) Are there any significant differences in data [censored] between geometries J, T, A (i.e., juveniles, teenagers, adults)?

An analysis of variance (ANOVA) was performed for both original and lognormally transformed data. At the 95% confidence level there were significant differences only as noted below:

	Raw	Log-Transformed
Enewetak & Medrin	A > T	A > (J,T)
Majatto	A > T	A > (J,T)
Ebeye	-----*	---------
Majuro	-----	---------
Utirik	-----	A > T

(* = no significant differences)

Figures 3a-e illustrate the distribution in activities measured in each chair at each island, separated by age distribution.

5. Question: Are there any significant differences in measurements in any one chair as indicated by recounts, or between chairs as indicated by crosscounts? [Using censored data.]

There were 16 recounting events, eight for each chair. For each event we calculated the percent difference in measurements, then rank ordered all 16 events. We then performed a linear regression in terms of rank. Figure 4 shows the results, with about as many differnces greater than zero were found than those which were less than zero, and where the only finding of possible significance is that the five largest differences, on the order of 20% or more (plus or minus), occurred in chair #2.

There were 25 events where crosscounts were made between chairs. Using the same type of rank ordering of differences described in the previous paragraph, about as many differnces greater than zero were found than those which were less than zero. Greatest differences (about + or - 20% or more) generally occurred at Ebeye and Majatto, although this is no more than a speculative observation.

TABLE 18 -- SUMMARY OF MARSHALL ISLANDS CS-137 DATA FOR EACH CHAIR

ENEWETAK				MAJATTO				EBEYE				MAJURO				UTIRIK			
CHAIR #1	Full		Censored*	CHAIR #1	Full		Censored	CHAIR #1	Full		Censored	CHAIR #1	Full		Censored	CHAIR #1	Full		Censored
Count	107	Count	95	Count	47	Count	19	Count	71	Count	6	Count	131	Count	62	Count	101	Count	101
Average	0.0142	Average	0.0162	Average	0.0025	Average	0.0033	Average	0.0022	Average	0.0059	Average	0.0072	Average	0.0129	Average	0.0369	Average	0.0369
Std Dev	0.0159	Std Dev	0.0162	Std Dev	0.0015	Std Dev	0.0022	Std Dev	0.0016	Std Dev	0.0038	Std Dev	0.0111	Std Dev	0.0140	Std Dev	0.0190	Std Dev	0.0190
Var	0.0003	Var	0.0003	Var	0.0000	Var	0.0000	Var	0.0000	Var	0.0000	Var	0.0001	Var	0.0002	Var	0.0004	Var	0.0004
High	0.0907	High	0.0907	High	0.0118	High	0.0118	High	0.0120	High	0.0120	High	0.0602	High	0.0602	High	0.0909	High	0.0909
90%	0.0907	90%	0.0530	90%	0.0035	90%	0.0058	90%	0.0021	90%	0.0120	90%	0.0249	90%	0.0336	90%	0.0658	90%	0.0658
Median	0.0093	Median	0.0108	Median	0.0020	Median	0.0027	Median	0.0019	Median	0.0054	Median	0.0022	Median	0.0060	Median	0.0335	Median	0.0335
10%	0.0021	10%	0.0026	10%	0.0018	10%	0.0021	10%	0.0018	10%	0.0021	10%	0.0020	10%	0.0025	10%	0.0175	10%	0.0175
Low	0.0012	Low	0.0023	Low	0.0018	Low	0.0018	Low	0.0018	Low	0.0021	Low	0.0019	Low	0.0021	Low	0.0037	Low	0.0037
CHAIR #2	Full		Censored	CHAIR #2	Full		Censored	CHAIR #2	Full		Censored	CHAIR #2	Full		Censored	CHAIR #2	Full		Censored
Count	121	Count	108	Count	53	Count	29	Count	76	Count	19	Count	134	Count	48	Count	100	Count	99
Average	0.0135	Average	0.0150	Average	0.0025	Average	0.0033	Average	0.0022	Average	0.0039	Average	0.0056	Average	0.0129	Average	0.0329	Average	0.0332
Std Dev	0.0179	Std Dev	0.0184	Std Dev	0.0013	Std Dev	0.0013	Std Dev	0.0026	Std Dev	0.0048	Std Dev	0.0108	Std Dev	0.0156	Std Dev	0.0181	Std Dev	0.0179
Var	0.0003	Var	0.0003	Var	0.0000	Var	0.0000	Var	0.0000	Var	0.0000	Var	0.0001	Var	0.0002	Var	0.0003	Var	0.0003
High	0.1097	High	0.1097	High	0.0056	High	0.0056	High	0.0214	High	0.0214	High	0.0549	High	0.0549				
90%	0.1097	90%	0.0490	90%	0.0048	90%	0.0055	90%	0.0214	90%	0.0125	90%	0.0134	90%	0.0430	High	0.0956	High	0.0956
Median	0.0083	Median	0.0097	Median	0.0020	Median	0.0028	Median	0.0016	Median	0.0021	Median	0.0018	Median	0.0056	90%	0.0602	90%	0.0594
10%	0.0015	10%	0.0085	10%	0.0012	10%	0.0019	10%	0.0013	10%	0.0016	10%	0.0014	10%	0.0020	Median	0.0292	Median	0.0294
Low	0.0013	Low	0.0013	Low	0.0011	Low	0.0012	Low	0.0013	Low	0.0013	Low	0.0013	Low	0.0014	10%	0.0140	10%	0.0141
																Low	0.0014	Low	0.0048

* Censored data are those above MDL

TABLE 1A -- SUMMARY OF MARSHALL ISLANDS CS-137 DATA

ENEWETAK				MAJATTO				EBEYE				MAJURO				UTIRIK			
	Full		Censored*		Full		Censored		Full		Censored		Full		Censored		Full		Censored
Count	228	Count	203	Count	100	Count	48	Count	147	Count	25	Count	265	Count	110	Count	201	Count	200
Average	0.0139	Average	0.0153	Average	0.0025	Average	0.0033	Average	0.0022	Average	0.0044	Average	0.0064	Average	0.0129	Average	0.0349	Average	0.0351
Std Dev	0.0170	Std Dev	0.0174	Std Dev	0.0014	Std Dev	0.0017	Std Dev	0.0022	Std Dev	0.0047	Std Dev	0.0110	Std Dev	0.0147	Std Dev	0.0187	Std Dev	0.0185
Var	0.0003	Var	0.0003	Var	0.0000	Var	0.0000	Var	0.0000	Var	0.0000	Var	0.0001	Var	0.0002	Var	0.0003	Var	0.0003
High	0.1097	High	0.1097	High	0.0118	High	0.0118	High	0.0214	High	0.0215	High	0.0602	High	0.0602	High	0.0956	High	0.0956
90%	0.0279	90%	0.0331	90%	0.0044	90%	0.0055	90%	0.0021	90%	0.0125	90%	0.0215	90%	0.0416	90%	0.0628	90%	0.0628
Median	0.0086	Median	0.0097	Median	0.0020	Median	0.0027	Median	0.0018	Median	0.0022	Median	0.0021	Median	0.0056	Median	0.0318	Median	0.0303
10%	0.0021	10%	0.0030	10%	0.0013	10%	0.0021	10%	0.0014	10%	0.0015	10%	0.0014	10%	0.0022	10%	0.0158	10%	0.0160
Low	0.0012	Low	0.0013	Low	0.0011	Low	0.0012	Low	0.0013	Low	0.0013	Low	0.0013	Low	0.0014	Low	0.0014	Low	0.0037

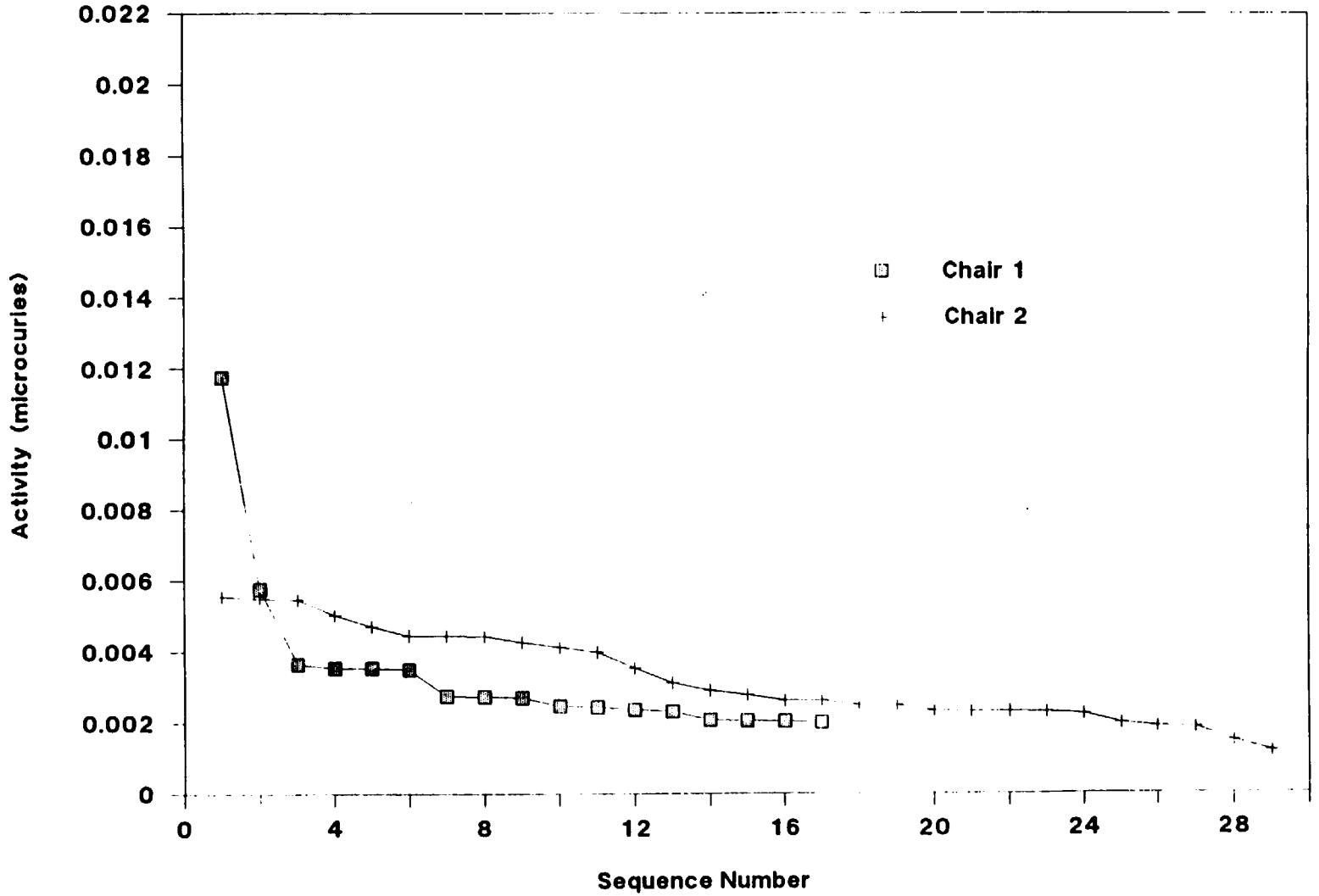
* Censored data are those above MDL

TABLE 2 -- COMPARISON OF SHAPE OF CS-137 DISTRIBUTIONS

	ENEWETAK	MAJATTO	EBEYE	MAJURO	UTIRIK
Original Data					
Mean	0.016	0.003	0.004	0.014	0.035
Std. Dev.	0.018	0.002	0.005	0.015	0.019
Median	0.010	0.003	0.002	0.006	0.030
Skewness	3.009	2.790	2.405	1.413	0.944
S.E. Skew	0.174	0.350	0.464	0.238	0.172
Kurtosis	10.613	11.738	6.104	0.852	0.454
S.E. Kurt	0.346	0.688	0.902	0.472	0.342
LOGNORMAL Data					
Mean	-4.532	-5.807	-5.786	-4.890	-3.497
Std. Dev.	0.862	0.421	0.763	1.079	0.570
Median	-4.585	-5.915	-6.119	-5.150	-3.497
Skewness	0.244	0.703	1.293	0.429	-0.617
S.E. Skew	0.174	0.350	0.464	0.238	0.172
Kurtosis	0.141	1.216	0.567	-1.153	1.082
S.E. Kurt	0.346	0.688	0.902	0.472	0.342

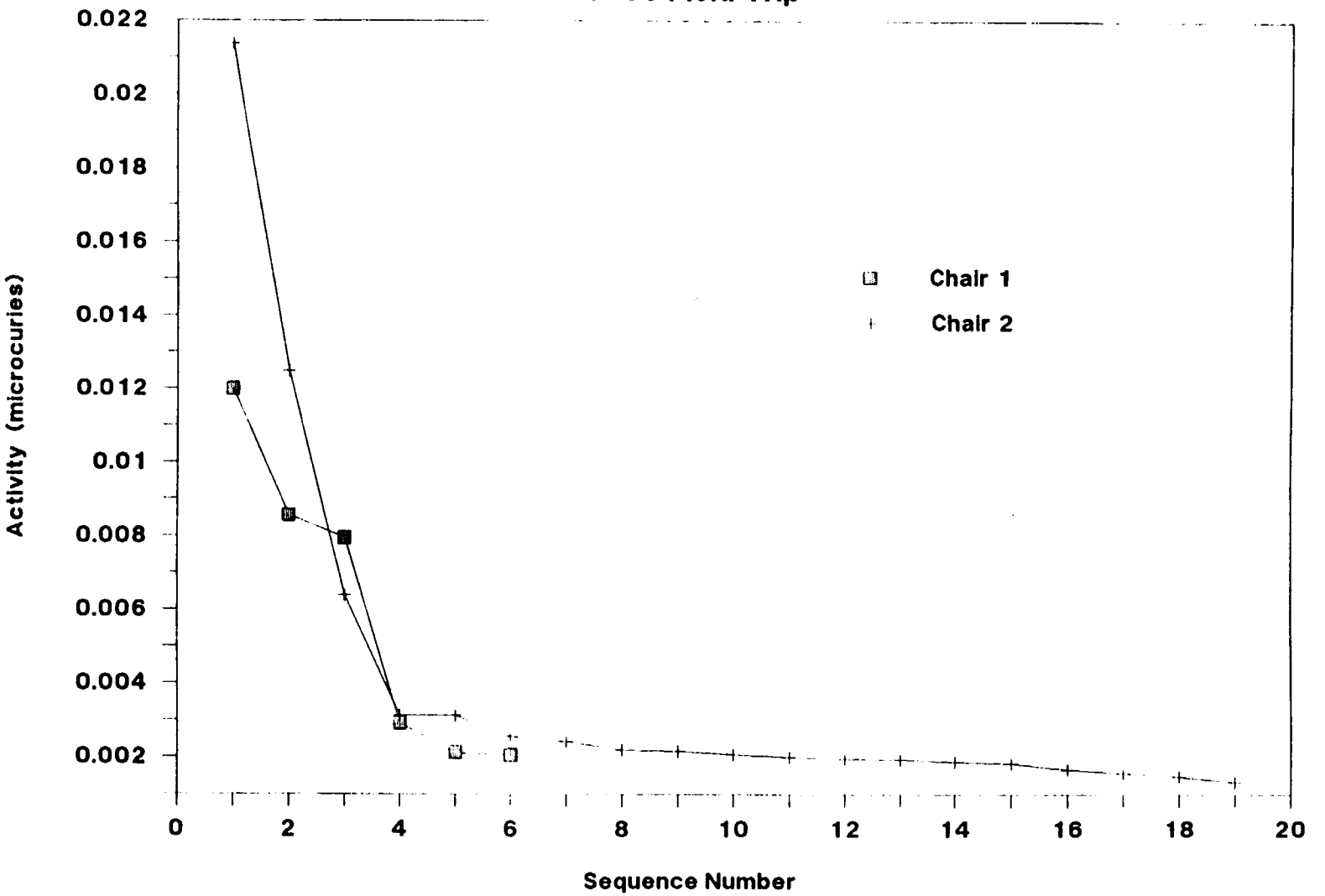
Cs137 Activity for Majatto

1989 Field Trip



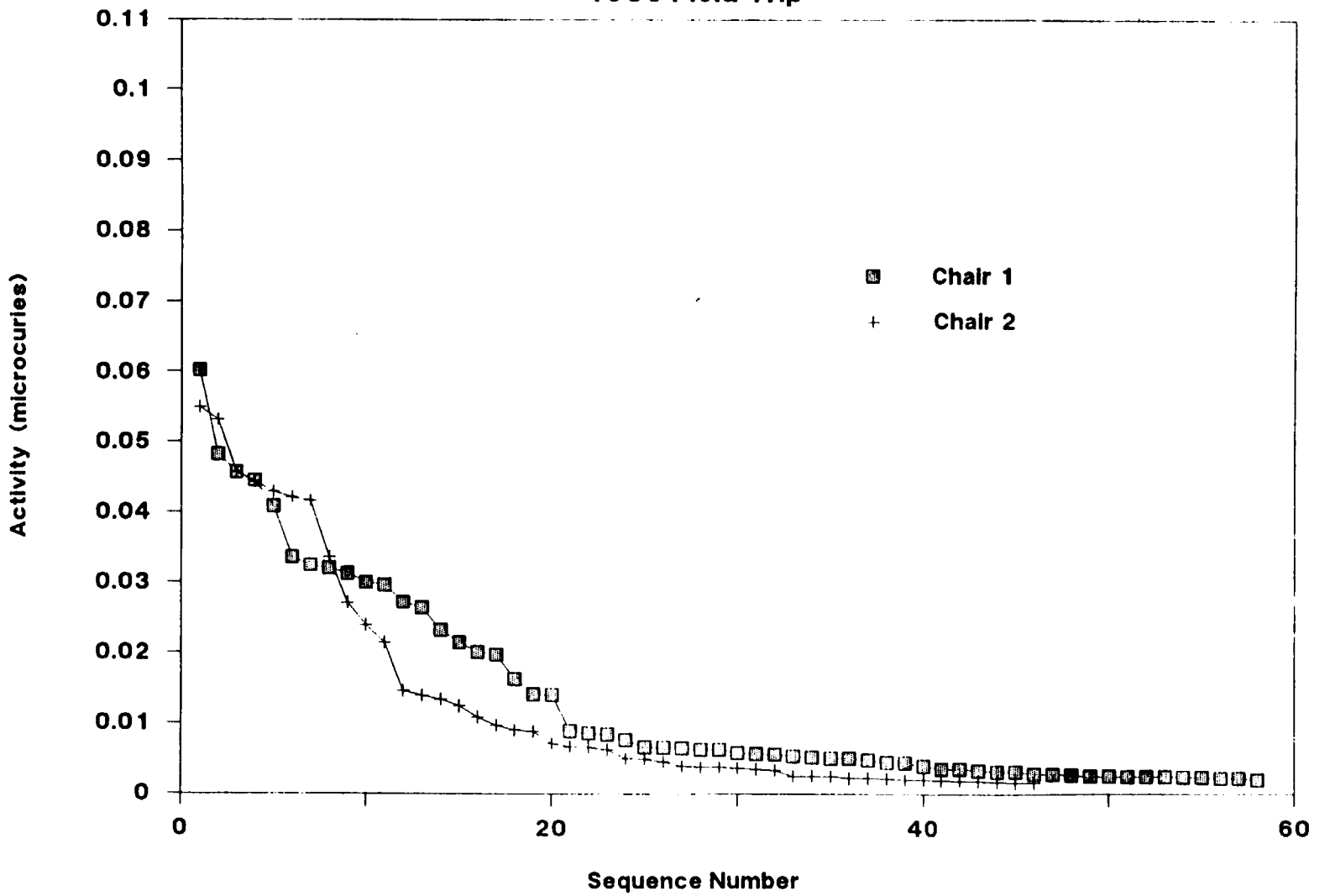
Cs137 Activity for Ebeye

1989 Field Trip



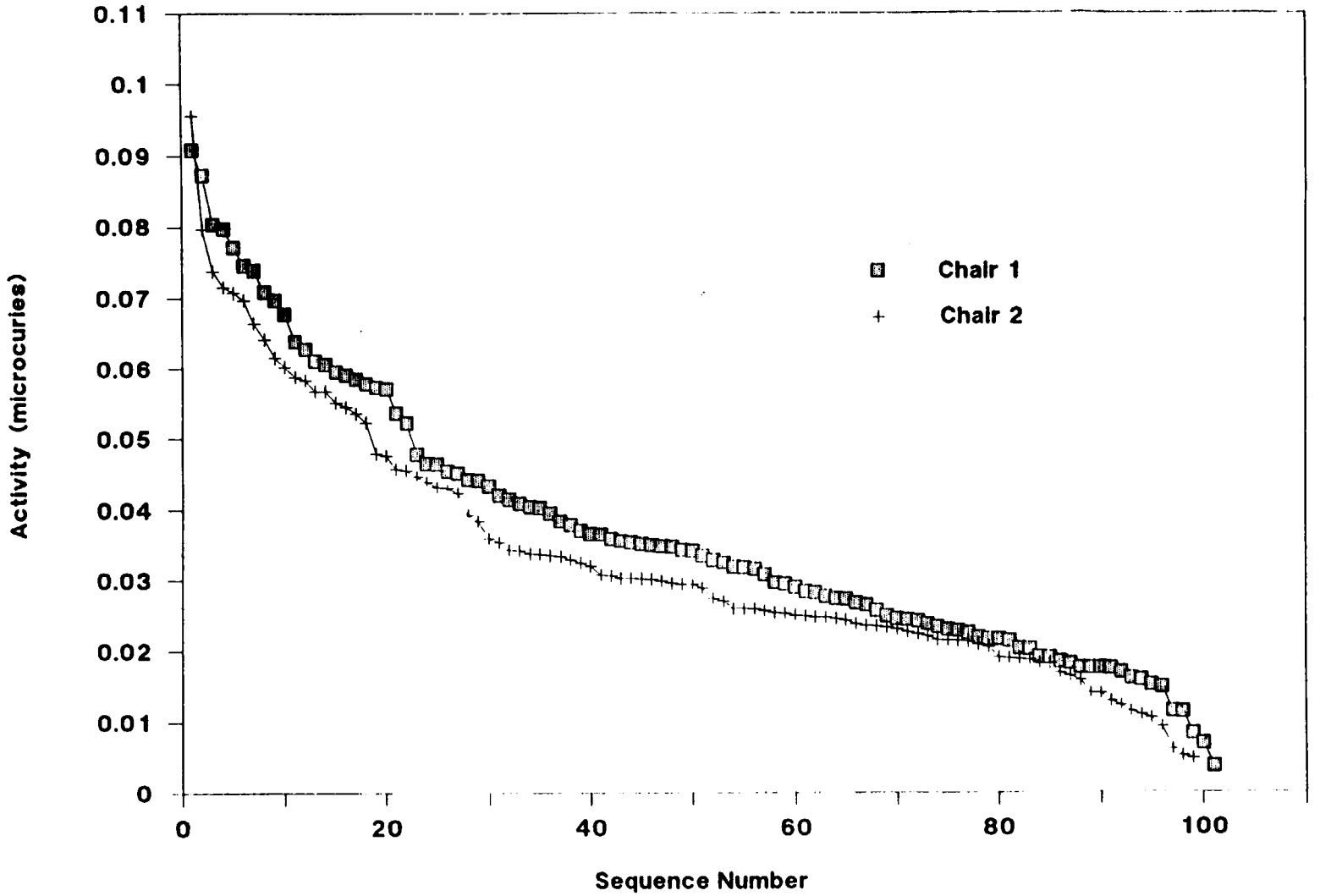
Cs137 Activity for Majuro

1989 Field Trip



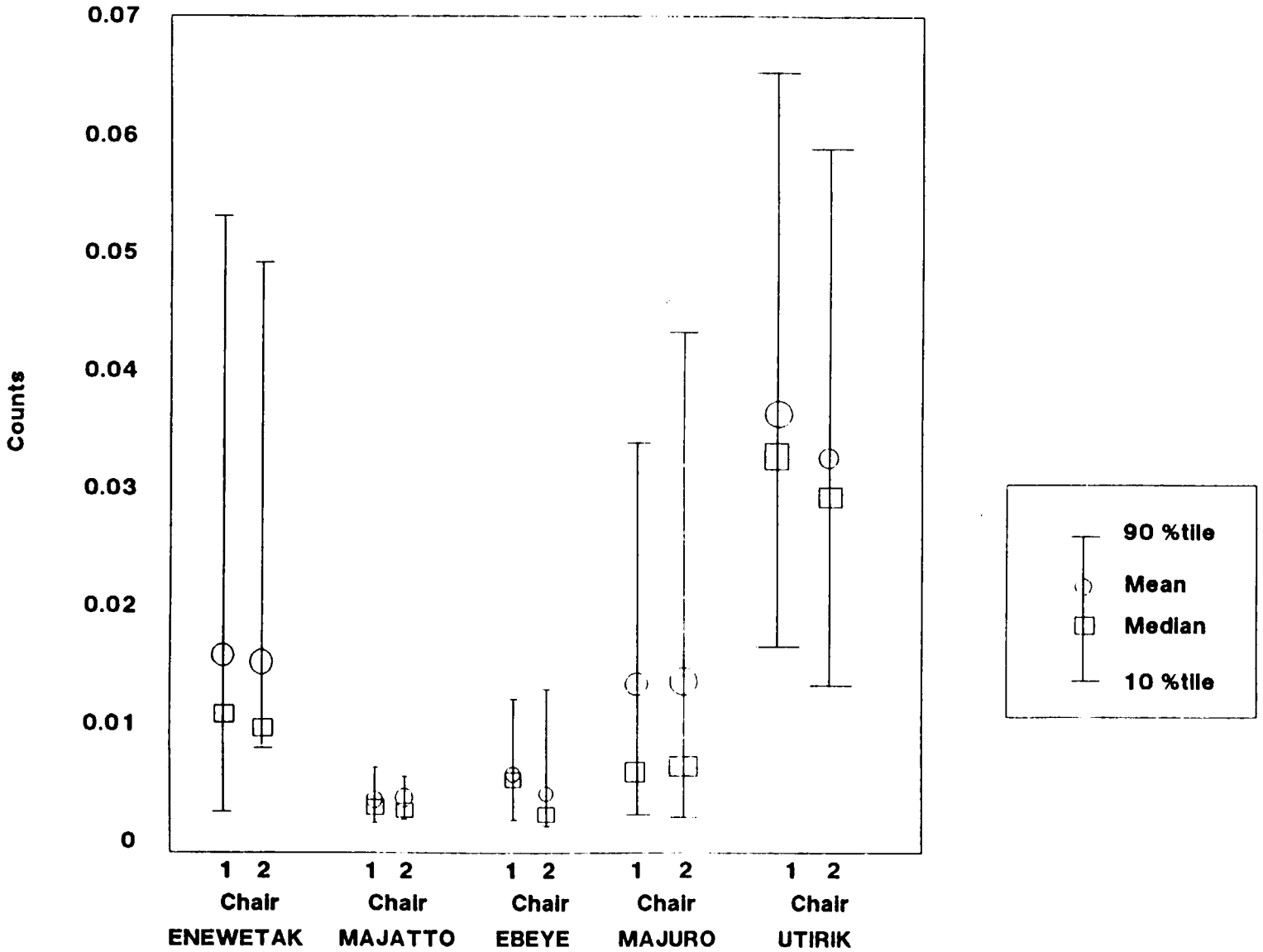
Cs137 Activity for Utirik

1989 Field Trip



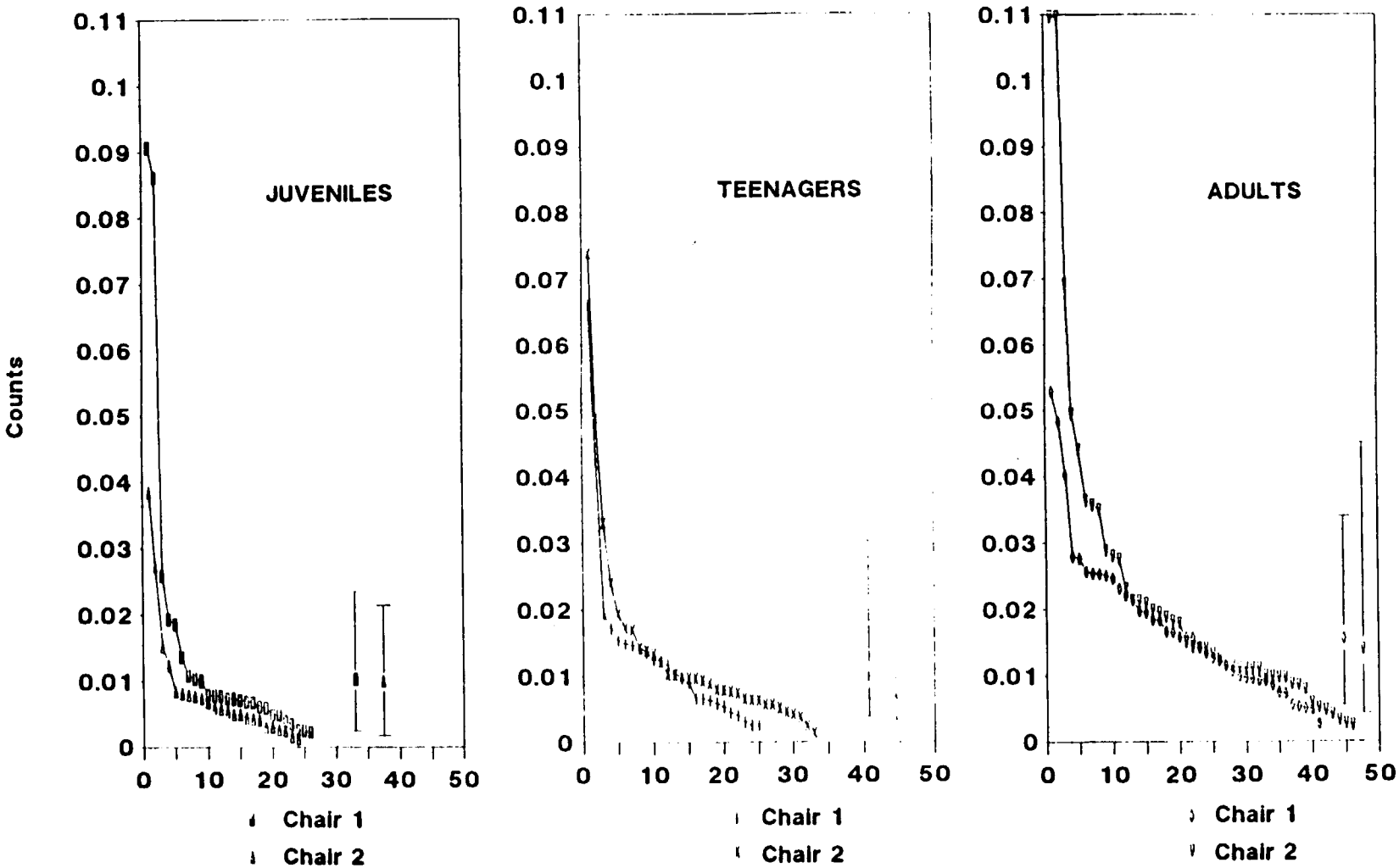
DRAFT
11/16/89

Summary of Whole Body Counts



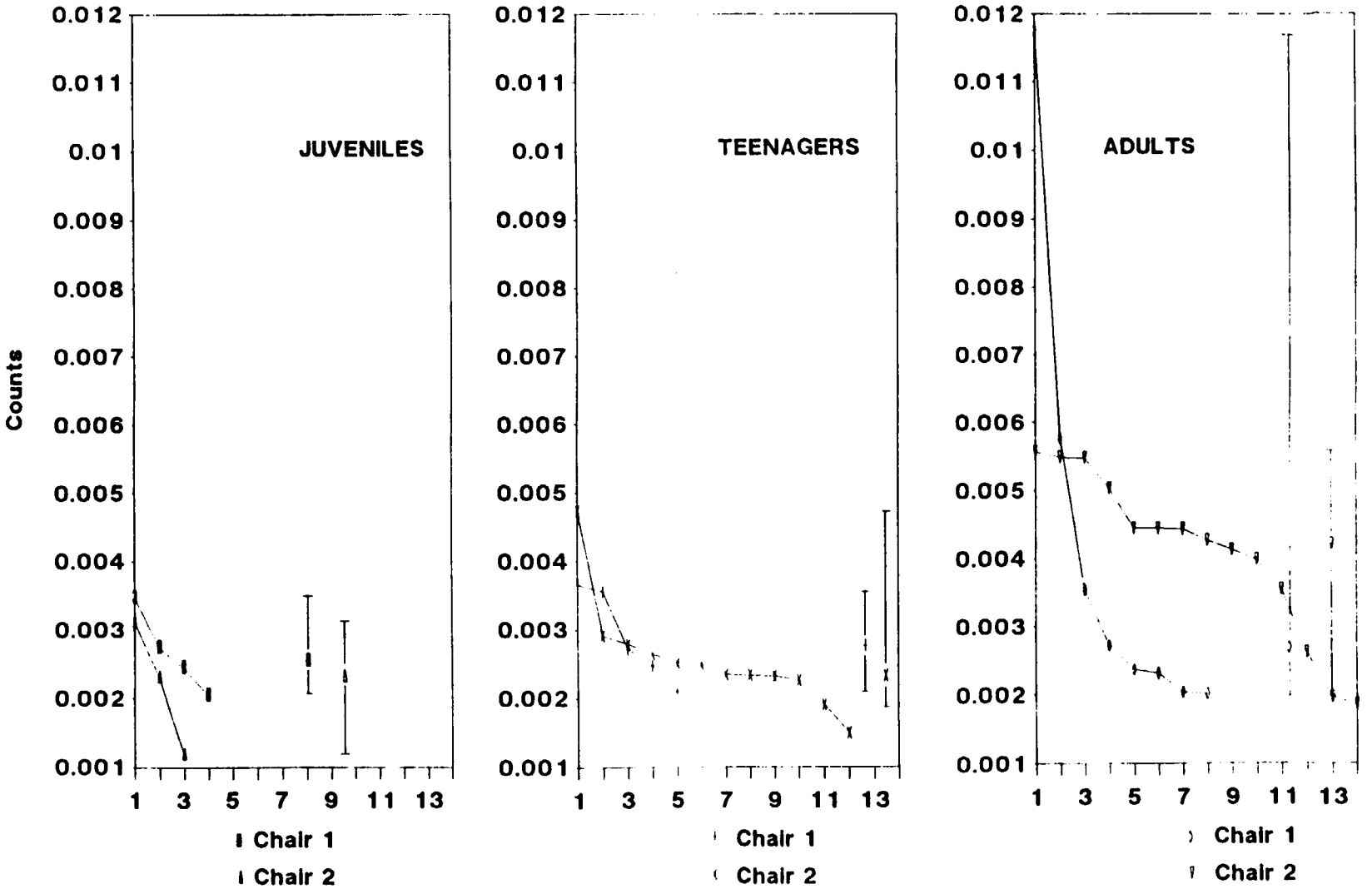
ENEWETAK AND MEDRIN

(Chair 1: 0.003-0.011-0.053; Chair 2: 0.008-0.010-0.049)



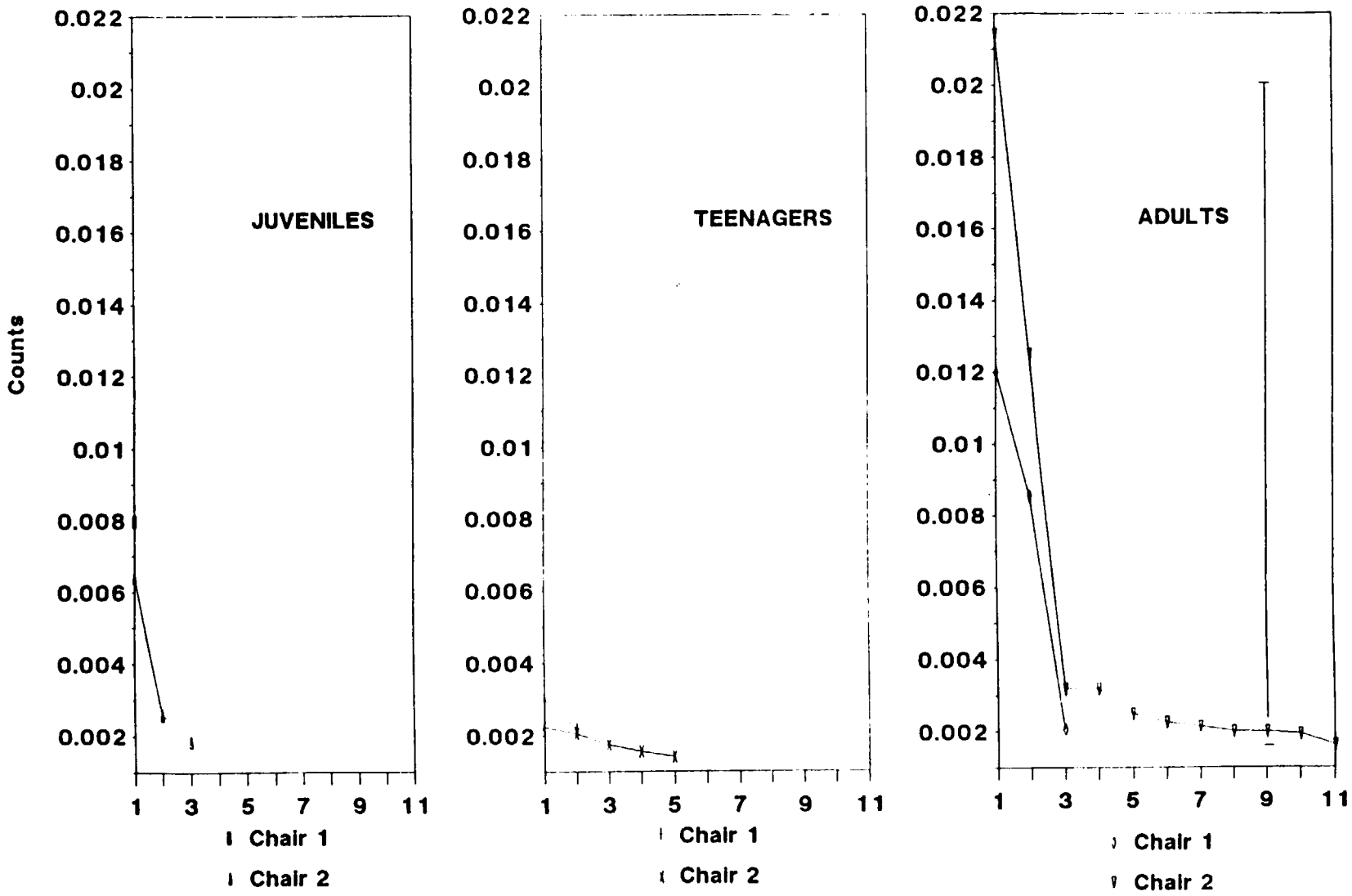
MAJATTO

(Chair 1: 0.002-0.003-0.006; Chair 2: 0.002-0.003-0.006)



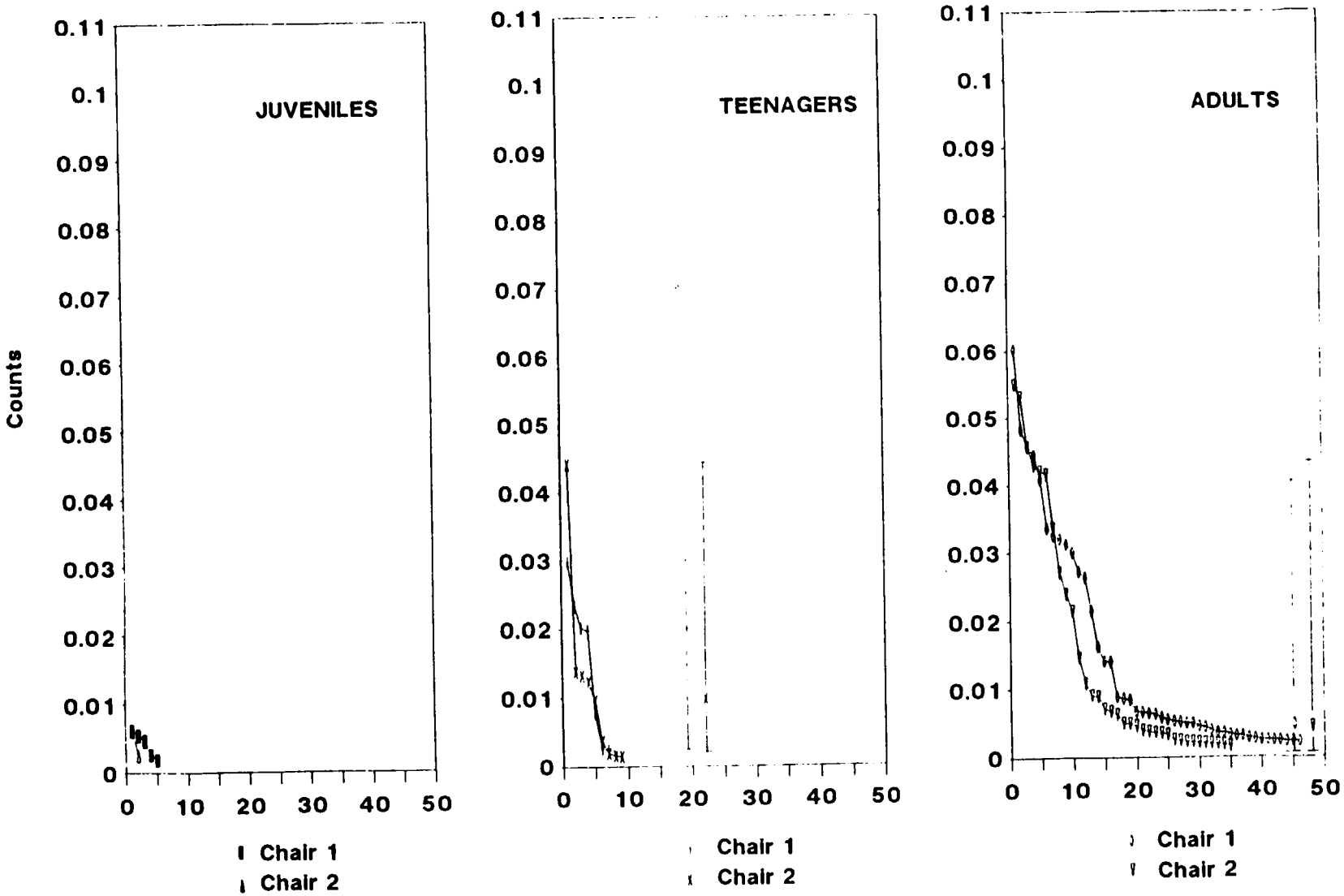
Ebeye

(Chair 1: 0.002-0.005-0.012; Chair 2: 0.002-0.002-0.012)



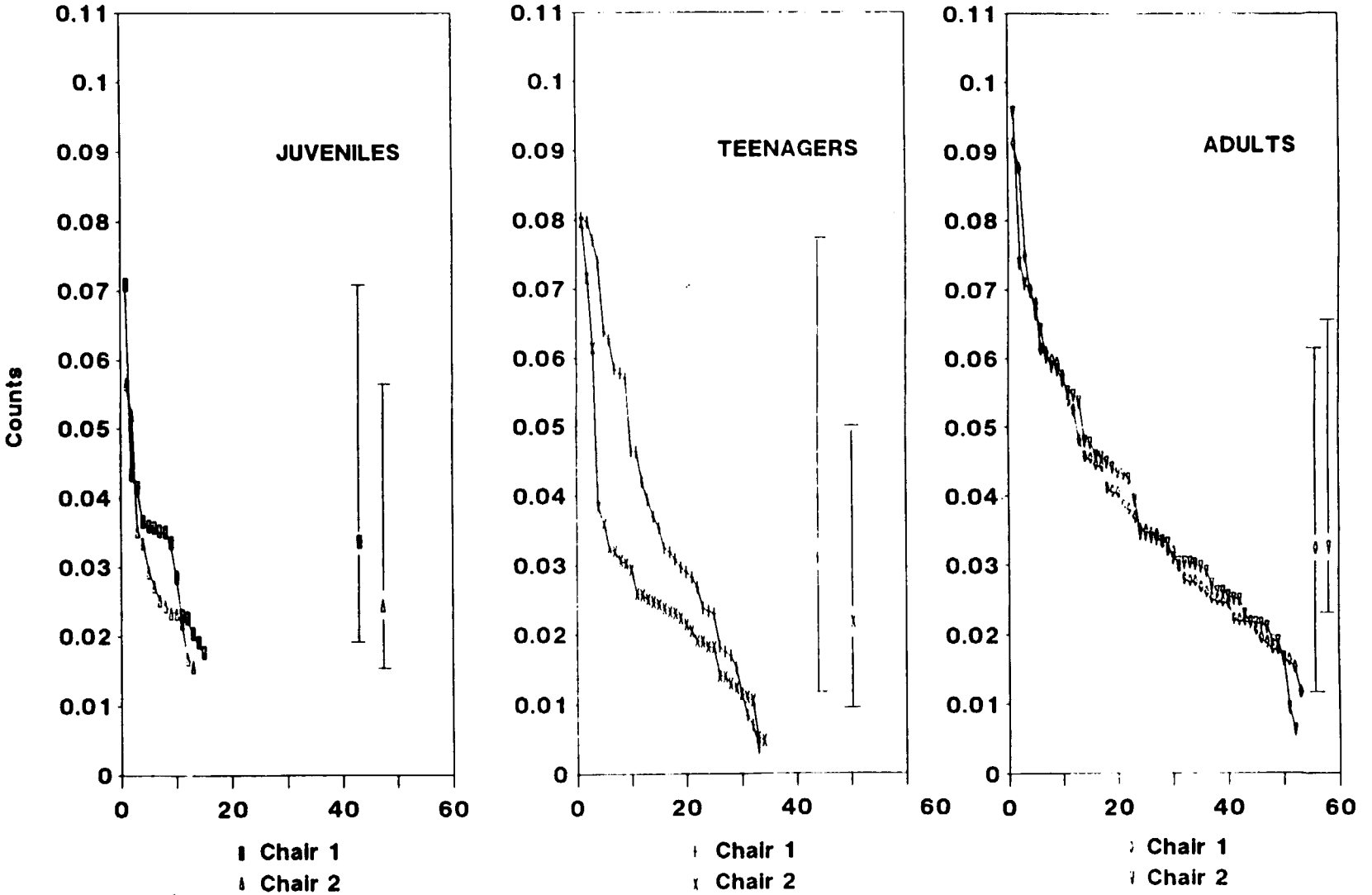
Majuro

(Chair 1: 0.002-0.006-0.034; Chair 2: 0.002-0.006-0.043)

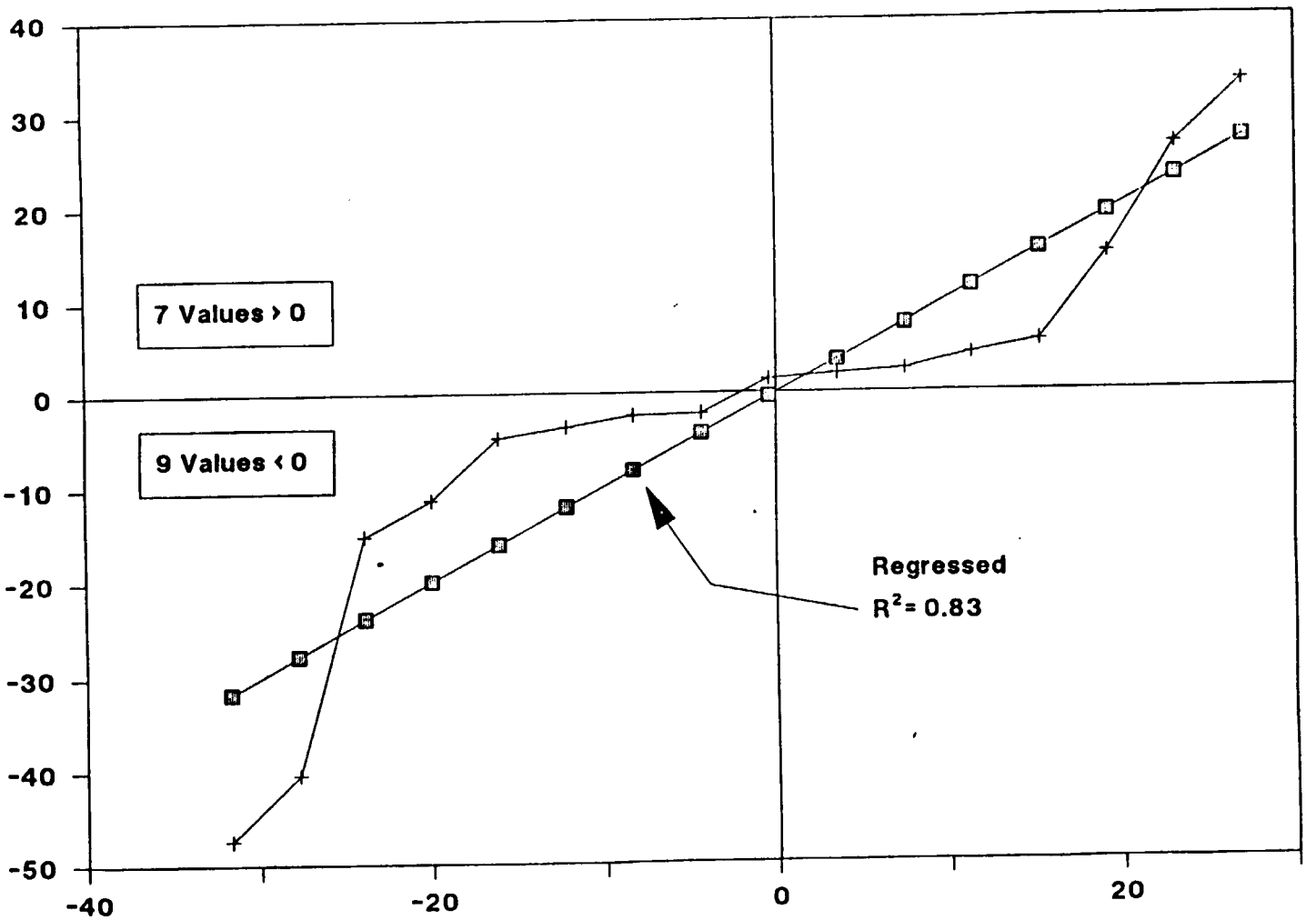


Utirik

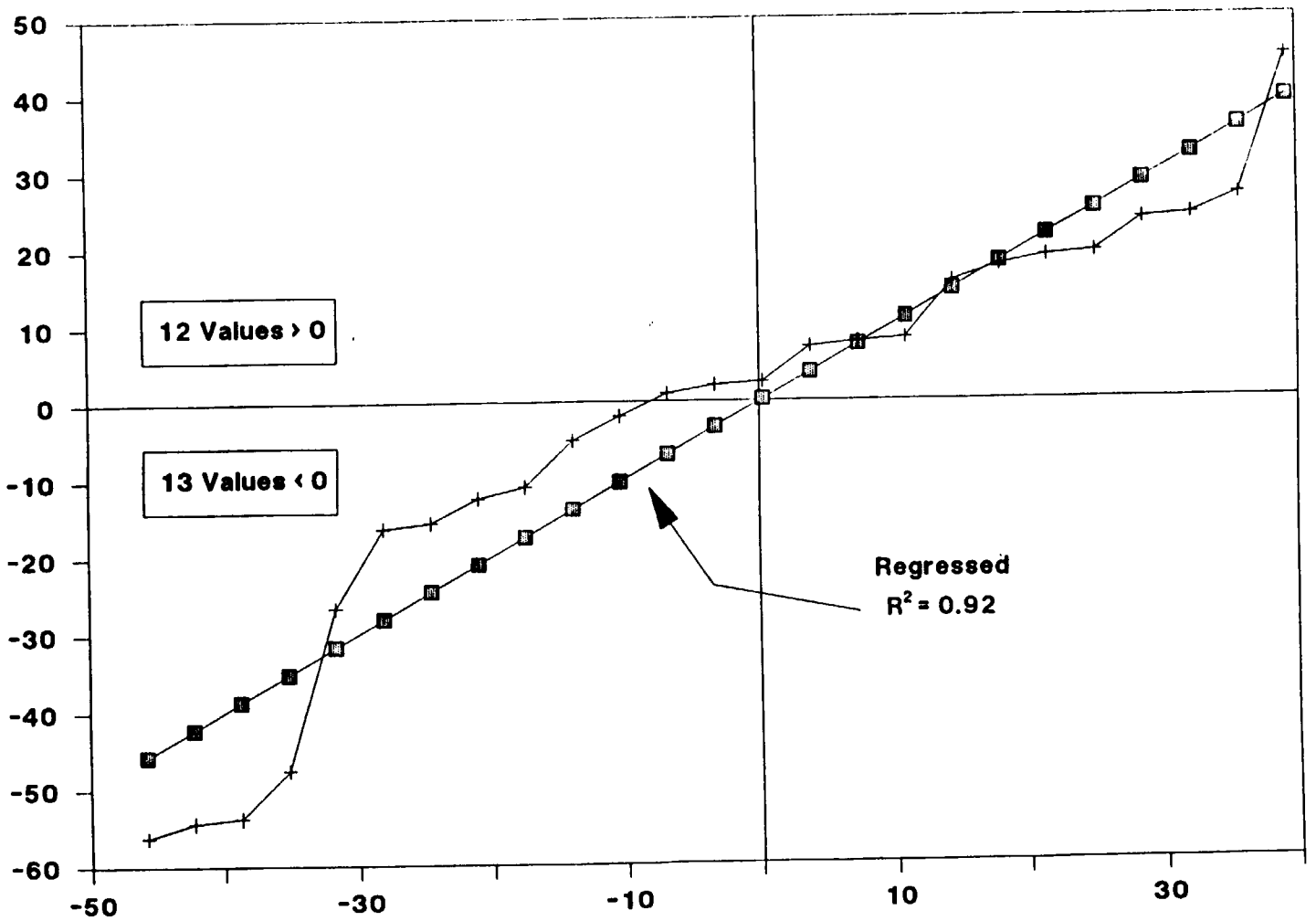
(Chair 1: 0.018-0.034-0.066; Chair 2: 0.014-0.029-0.059)



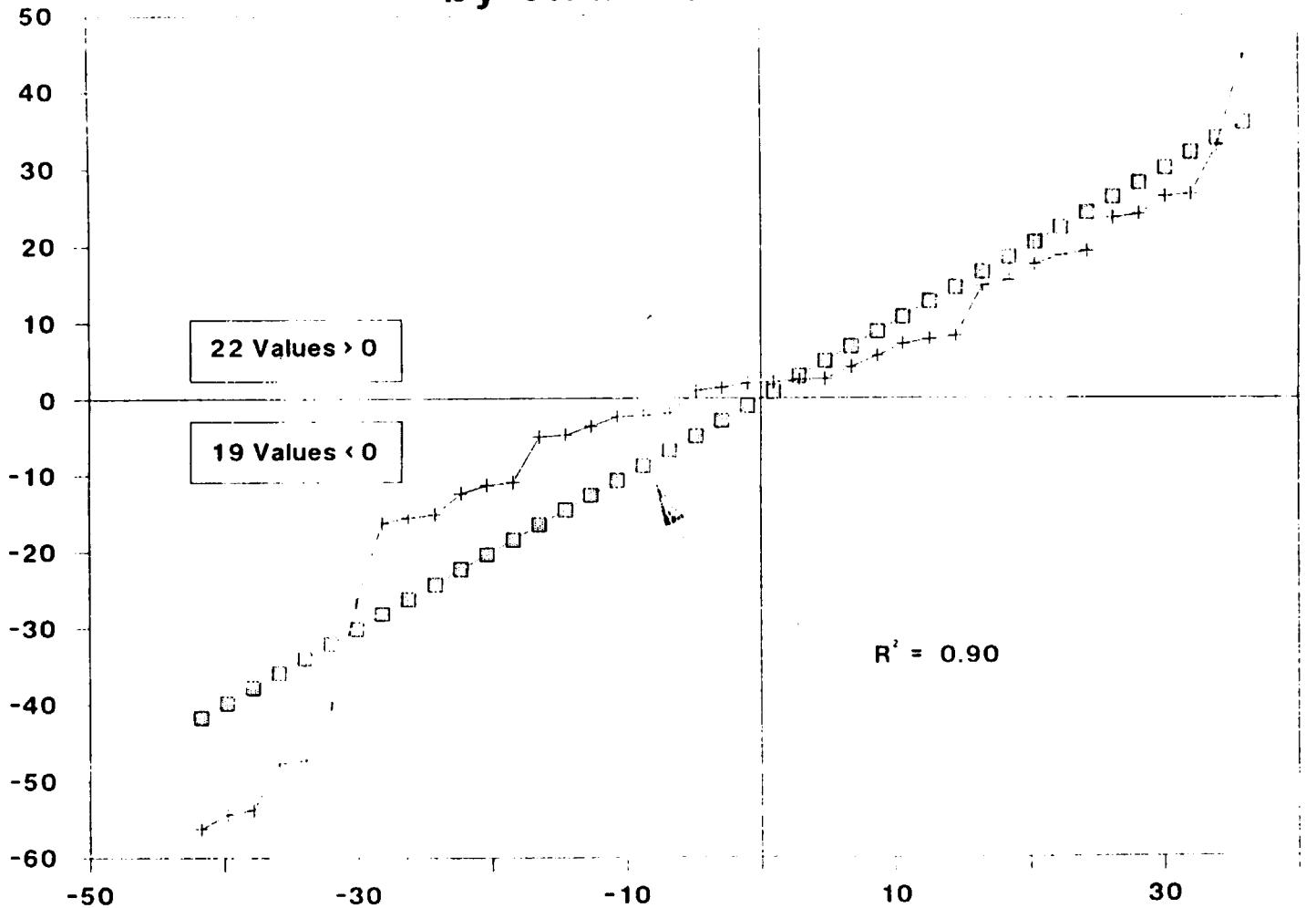
Differences in Cs-137 Recounts



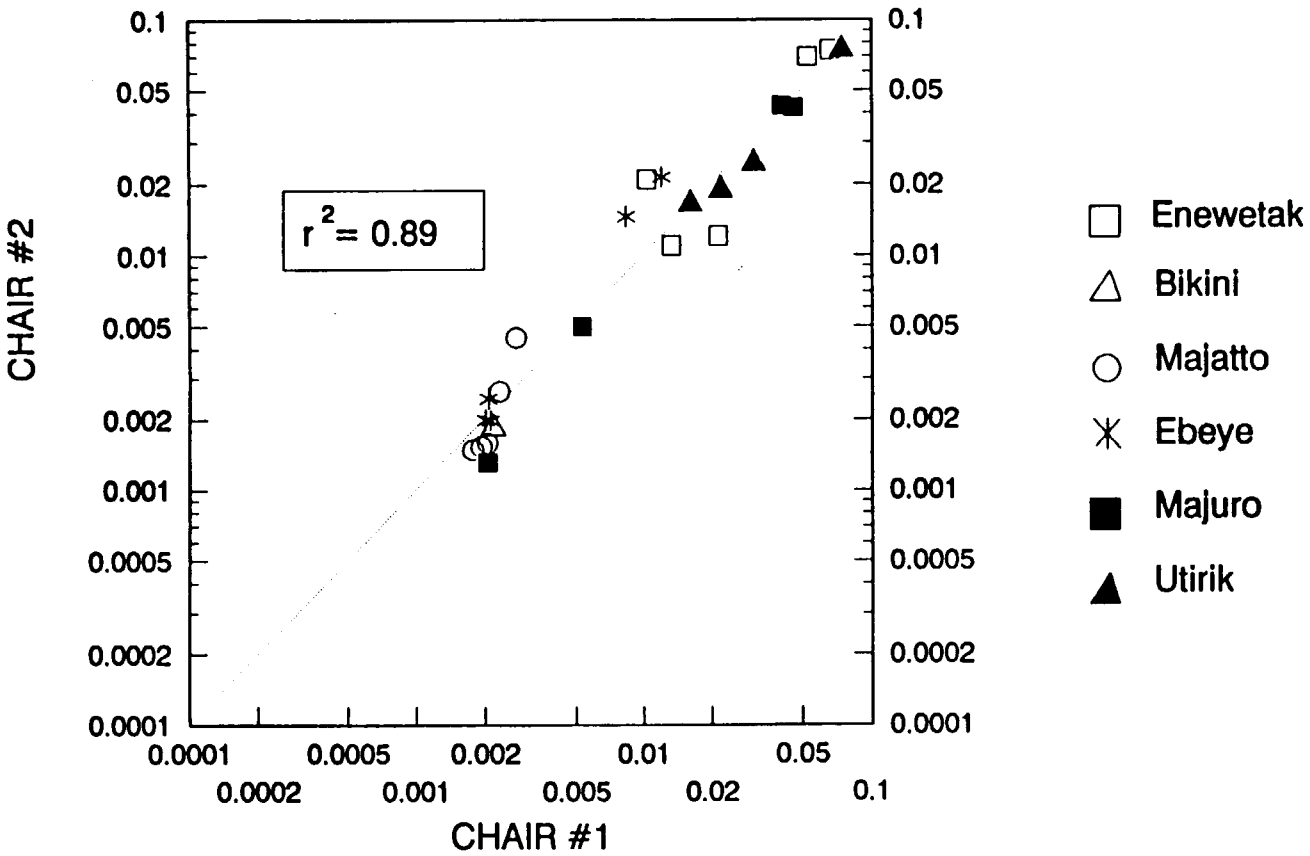
Differences in Cs-137 Crosscounts



Differences in Cs-137 as Measured by Chairs 1 and 2



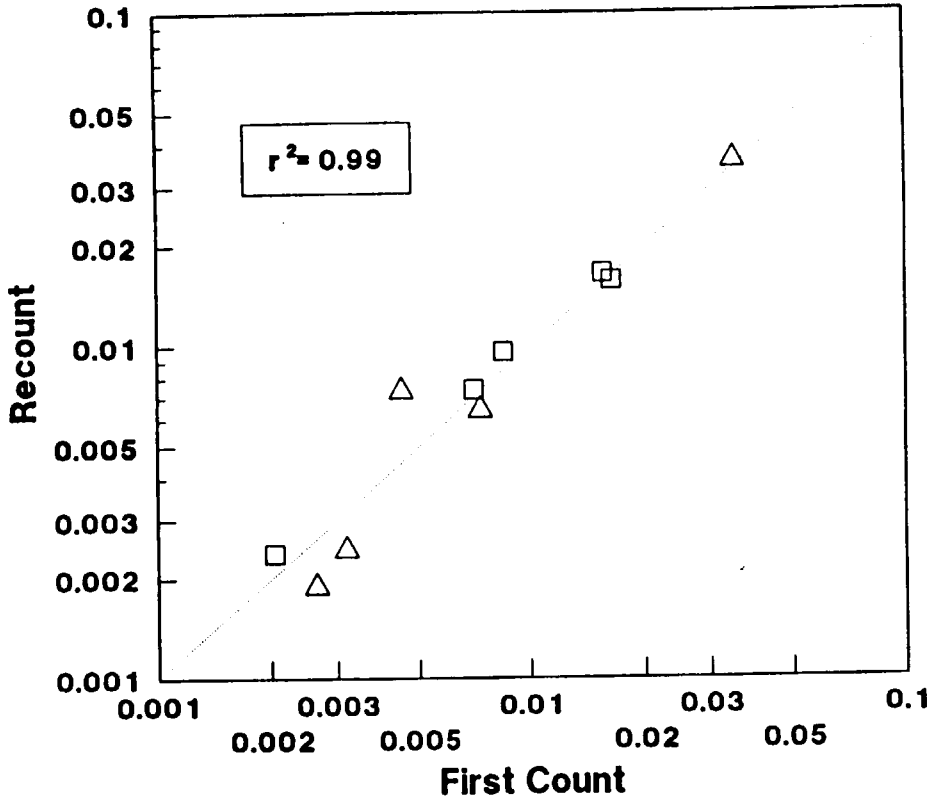
Marshall Island Cross Count Data (1989)



2/27/90

Marshall Island Recount Data for Chair #1

(1989)



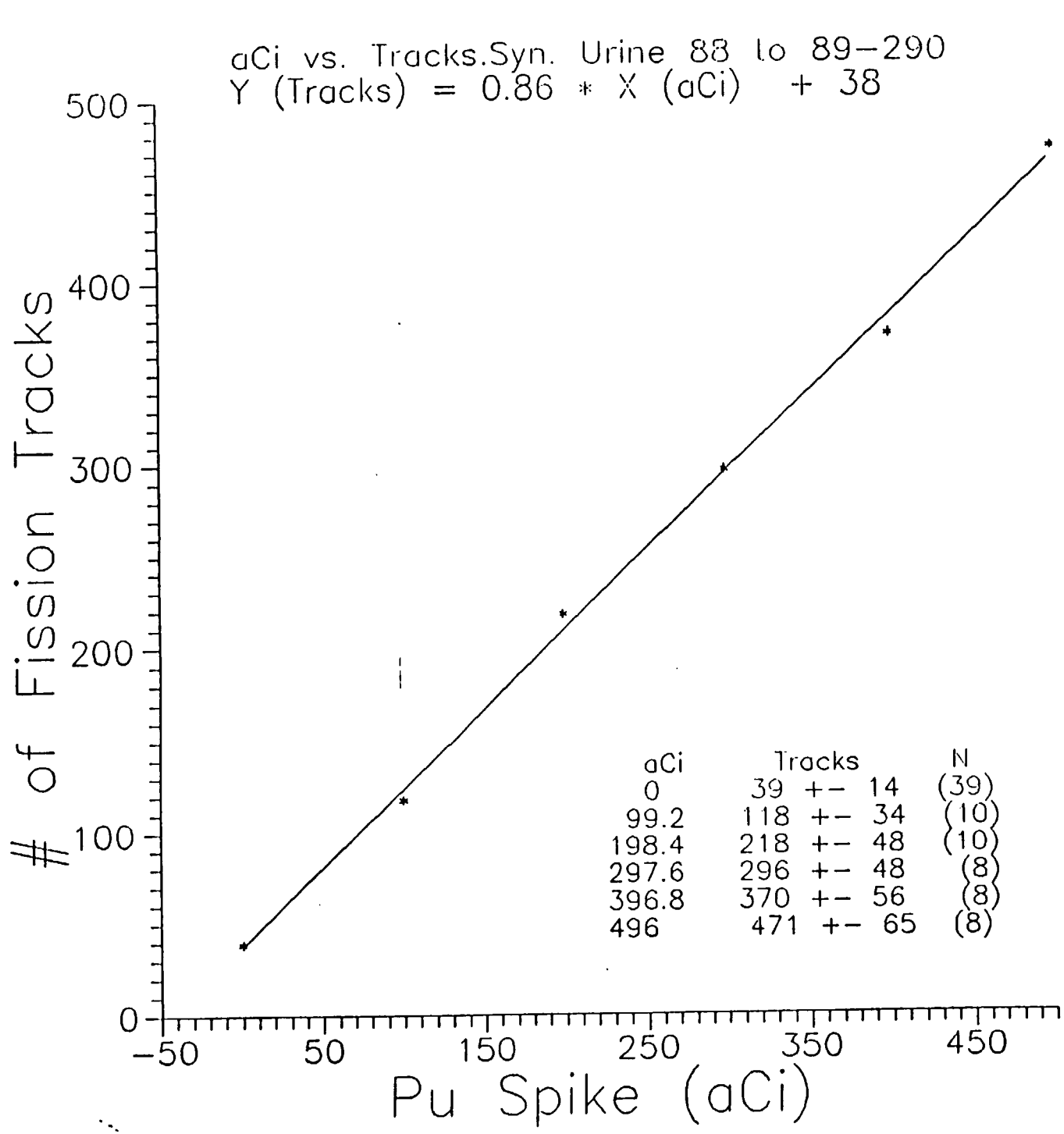
2/27/90

Quality Assurance Program

The attached protocol defines the quality control of the fission track analysis method. As described, sixteen samples are prepared for analyses, in two batches of eight. Three synthetic urine samples are spiked, one at 100 aCi, the second at 500 aCi and the third varying between these two values. In addition two synthetic urine blanks, one split and ten other urine samples of the Marshallese, totaling 16, are processed for FTA.

A QC chart is maintained to ascertain that the results of each 500 aCi spike falls within the 2 sigma of an expected value derived from the average of the previous 10 measurements of synthetic urine spiked at a level of 500 aCi. If the recovery falls outside the 2 sigma values, continuation of further Pu analysis is contingent upon Dr. Moorthy's written approval. This enables us to monitor any recovery problems or contamination.

Figure shows the track vs. aCi spike derived from the spiked urines and blanks. Such a relationship is used to convert the tracks from the urine samples to aCis.


aCi vs. Tracks.Syn. Urine 88 to 89-290
Y (Tracks) = 0.86 * X (aCi) + 38
# of Fission Tracks
Pu Spike (aCi)
0
100
200
300
400
500
-50
50
150
250
350
450
aCi Tracks N
0 39 +- 14 (39)
99.2 118 +- 34 (10)
198.4 218 +- 48 (10)
297.6 296 +- 48 (8)
396.8 370 +- 56 (8)
496 471 +- 65 (8)

BNL/FTA

Sensitivity below 100 aCi/sample

Well Established QA program:

10 Marshallese Urine
3 Spiked Urine Samples
2 Urine-Blank Samples
+ 1 Split Sample

16

COMPARISON OF SPIKED URINE DATA

	1987			1989		
Y \ X	SIZE	MEAN	SIG	SIZE	MEAN	SIG
1000 aCi	4	592	51			
500 aCi	17	320	125	5	453	41
400 aCi	20	228	90	7	381	38
300 aCi	21	174	46	7	286	47
200 aCi	21	134	37	10	209	17
100 aCi	21	88	31	8	116	14
BLANKS	76	54	29	26	42	13

$Y=1.8(X-30)$ $Y=1.18(X-36)$

Plutonium from Atmospheric Weapons Testing: Fission Track Analysis of Urine Samples

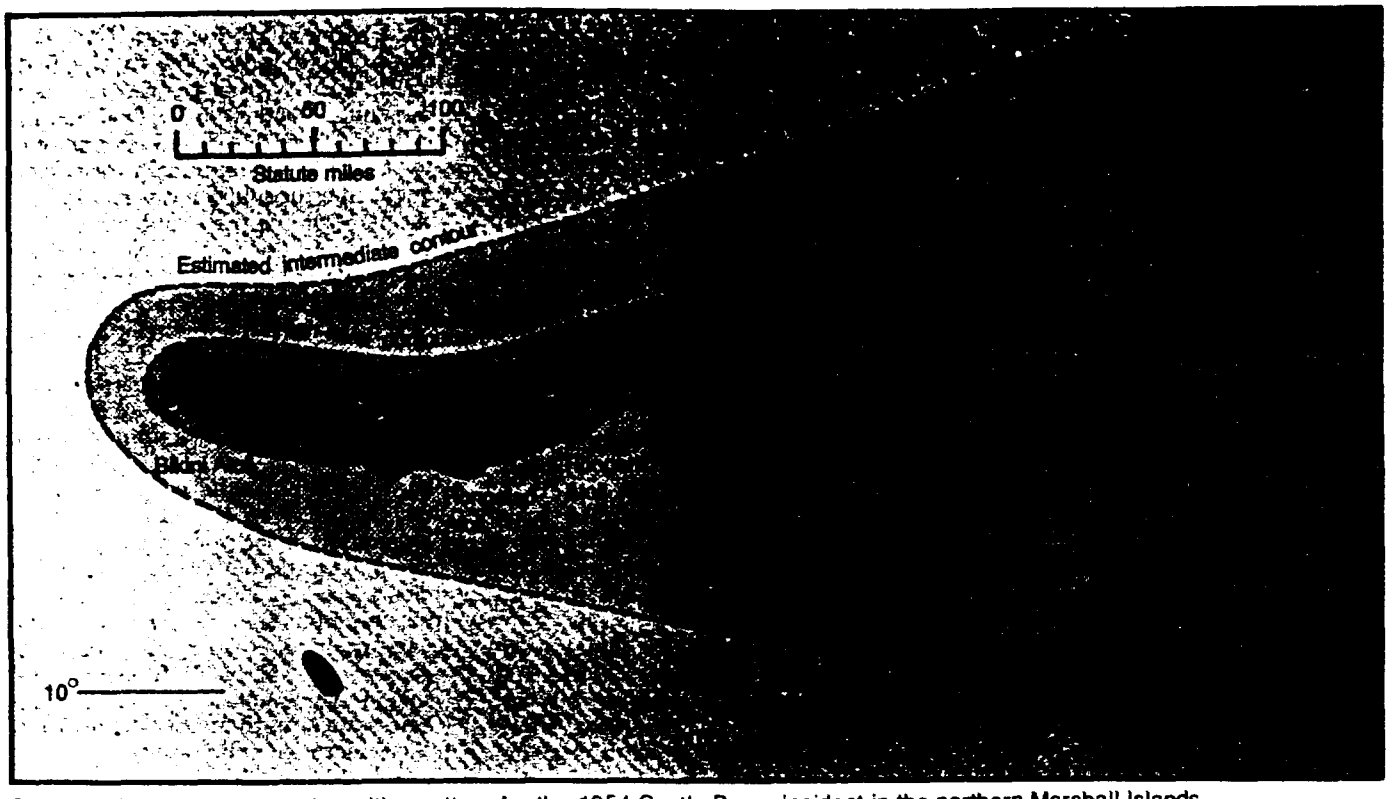


Figure 1. Schematic fallout deposition pattern for the 1954 Castle Bravo incident in the northern Marshall Islands.

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ANALYTICAL APPROACH

Between 1946 and 1958 the United States carried out a series of more than 60 atmospheric tests of nuclear weapons in the northern Marshall Islands. During the largest of these in March 1954, an unanticipated wind shift led to extensive surface contamination of inhabited atolls up to 500 km east of Bikini within a 5000-km^2 area (Figure 1). The health and radiobiological status of residents potentially exposed to the fallout has since been continuously monitored (*1*).

Analysis of Marshallese urine samples for ^{239}Pu was started at Brookhaven in 1983 after preliminary work indicated that urine composites from residents of Bikini contained about 45 fCi (1.7 millibecquerels)/L of α-activity in the ^{239}Pu region, as determined by Photon Electron Rejection Alpha Liquid Scintillation (PERALS) Counting. However, reanalysis with α-spectroscopy using surface barrier detectors revealed that most of the activity originated from ^{210}Po, whose α-spectrum overlaps with that of ^{239}Pu.

^{239}Pu, unlike ^{210}Po, undergoes thermal neutron-induced fission and can be determined by fission track analysis (FTA) (*2*). FTA is a type of neutron activation analysis whereby the analyte is placed on a detector and bombarded with neutrons, usually in a nuclear reactor. Upon fission, two fragments are released in opposite

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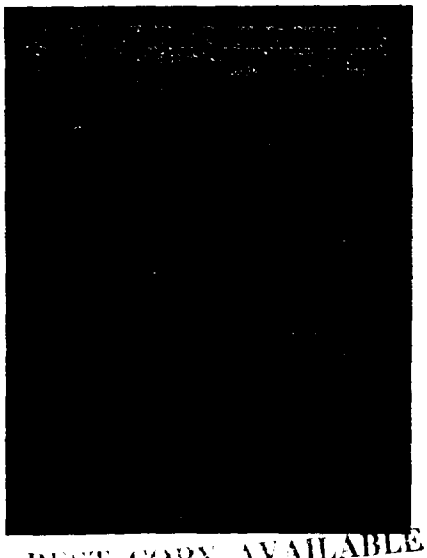
directions. One impinges on the detector—an insulating solid such as a crystal, polymer, or glass—and leaves a highly localized track that can then be counted.

The phenomenon was first reported in 1958 by Young (*3*), and fundamentally the technique has remained unchanged. Developments have been centered around improvements in the purity of detector materials (in order to minimize background tracks) and in track-counting methodology. With proper selection of detector and track revelation procedures, one can discriminate between different types of particles and determine particle parameters such as charge, energy, and mass.

The sensitivity of the procedure is a function of the number of fissionable events, which increases with the fission cross section of the nuclide, the neutron density, and the irradiation time. One drawback of FTA is that it does not distinguish between fissionable materials. For ^{239}Pu, the major interference is from U, which is ubiquitous in the environment. Our task, therefore, was to develop methodology for U decontamination so as to enable FTA to be applied to the determination of trace amounts of ^{239}Pu.

Summary of the procedure

The protocol used to isolate and determine ^{239}Pu is shown in the box. Ion-exchange isolation was done in two stages. The acid solution was placed on a 6-mL anion-exchange column, and the ^{239}Pu fraction was eluted in about 40 mL of eluent. Most of the salts in the sample were removed at this stage. It is important to eliminate dissolved solids because they could quench response by absorbing fission fragments. The eluate was concentrated and placed on a smaller (35-μL) column, and the ^{239}Pu was isolated in 200 μL of eluent. A 40-μL aliquot was then deposited on a fused-silica detector and irradiated with thermal neutrons. The resulting fission tracks were enlarged by chemical etching and then were optically counted.



Purification of materials

Acids were purified by sub-boiling rather than conventional distillation. Kuehner et al. (*4*) have shown that significant contamination of the distillate occurs in the latter method from creeping of the unrectified liquid and from entrainment of particulates in the vapor stream formed during bubble rupture. In sub-boiling distillation the acid is vaporized by gentle surface evaporation.

Uranium has been found at concentrations of 72 ppm in borosilicate glass (*4*) and 0.3 ppb in commercial quartz. Because U can be leached from glass by acids, quartz stills were used. For HCl, sub-boiling distillation in quartz reduced U contamination by a factor of 10, to 30–80 pptr. However, even a femtogram of natural U gives an average of 0.9 tracks upon irradiation under our conditions. To keep the track count below this level, it was necessary to reduce the natural U concentration to below 0.025 pptr in the 6 M HCl reagent used in the process. This was achieved by passing the HCl through an anion-exchange resin with a high affinity for U. No tracks above detector background were observed when 0.1 mL of the final product was evaporated and irradiated. Nitric acid (7.2 M) was similarly purified.

Critical stages of the technique were performed in a dust-free (Class 100) environment; U in ambient air, and particularly in dust particles, would otherwise have raised the background to intolerable levels.

Ion-exchange chromatography

The chromatography was intended to separate Pu from U and to isolate the former in a few microliters so that a small area of the detector could be conveniently spotted. The chromatography was conducted in two stages. Separation of Pu from salts present in the urine samples was accomplished in the first, and separation of Pu from U was achieved in the second.

The quartz column (11 × 0.8 cm i.d.) used in the first chromatographic stage was cleaned with hot HNO_3 for several days before use. Dowex-1 (50–100 mesh), precleaned to be free of resin fines, was transferred to the column and was further cleaned with HNO_3. The sample was then quantitatively transferred to the column, which was eluted sequentially with 7.2 M HNO_3, 6 M HCl, and a mixture of 6 M HCl and 0.1 M HI.

The eluate (40–45 mL) was transferred by weight into a quartz evaporation vessel that tapered to a 1-mL collection thimble. The solution was evaporated to dryness at 85 °C, redissolved in HNO_3, and treated in turn with $FeSO_4$ and $NaNO_2$ to ensure that the Pu was in the 4^+ state.

The second-stage chromatography was performed on a fused-silica microbore (4 × 0.7 mm i.d.) column in a Class 100 environment. The sample (200 μL) was transferred to the column, which was eluted sequentially with 7.2 M HNO_3, 6 M HCl, and a mixture of 6 M HCl and 0.008 M HF.

Initial recovery studies were conducted with about 10 pCi of ^{238}Pu because this isotope is an α-emitter and is much easier to monitor than ^{239}Pu. Recovery of Pu was >90% in the first stage; recovery from the second stage was much poorer (35%). The volume collected from the second column had to be kept to a minimum; in the subsequent step the solution was spotted on the detector and the liquid was evaporated. Hence the Pu band could not be fully collected, and a low recovery resulted. The overall recovery was about 30%. These values were confirmed with ^{239}Pu at the 1-fCi level.

Our chromatographic procedure differs from previously reported methods (*5*) in which Pu was eluted from Dowex-1 with 6–11 M HBr. This method was unsuitable for our purposes for two reasons. First, the reported decontamination factor from U (the ratio of U in the sample before and after treatment) was only about 3×10^5, and an unacceptably high background would have resulted. Second, the relatively good wetting ability of HBr would have spread the sample over the detector surface to a much greater degree than the eluent used in this study.

Selection and preparation of the detector

Fused silica is an excellent medium for recording high-energy particles such as fission fragments because it is relatively insensitive to incident particles of mass less than 40 amu with kinetic energies less than 100 MeV (*2*). Track images are retained indefinitely on normal storage.

The background of the detector is important in defining the detection limit. Detectors have been analyzed on numerous occasions by neutron activation analysis and microdot X-ray fluorescence and have been found to contain only parts-per-billion levels of inorganic contaminants and no measurable U. However, none of these techniques has the sensitivity necessary for measuring U at the femtogram level that is needed for the present investigation.

When concentrated HNO_3 and aqua regia were initially used in succession to clean the slides, a clustered nonuniform pattern of tracks resulted upon irradiation. This phenomenon was at-

Figure 2. Etched fission tracks at 100× magnification.

Figure 3. Track pattern from a U hot spot.

tributed to trace amounts of U-rich inclusions distributed on the top 5–10 μm of the detector during polishing. When concentrated aqua regia and hot nitric and sulfuric acids were used sequentially, the clusters were not observed. It is postulated that the inclusions were caused by the presence of trace amounts of U occluded by thorium oxides in the materials used for optical polishing. The problem disappeared in the presence of concentrated H_2SO_4, possibly because of dissolution of the thorium oxide whose solubility in sulfuric acid is higher than that in nitric acid.

Scratches on the slide resulting from optical polishing by the manufacturer interfered with the counting of fission tracks (*6*). These could be removed by fire polishing. However, a conventional brass torch proved unsatisfactory; sputtering of unidentified (probably metallic) particles from the torch contaminated the detector surface and led to a high background. Use of a gold-plated torch failed to resolve the problem, but a torch fabricated from fused silica—the same material used in the detector—gave good results.

Background tracks in fused silica from 50 observations averaged 4 ± 5 tracks per irradiation per cm^2 (number of thermal neutrons per cm^2 or fluence is 10^{17}/cm^2). Riley (*6*) used similar fused-silica detectors cleaned with methanol and dilute HNO_3. We found that this procedure led to a track density of 50 ± 70. Clearly, the elaborate cleaning and handling procedures developed in this study lead to much improved results.

Target preparation and irradiation

An aliquot of the sample solution (40 μL) was transferred to a 1-cm^2 area of the detector surface and evaporated to dryness under an infrared lamp in a Class 100 workstation. A known amount of ^{239}Pu was added to another part of the detector as a flux monitor and was similarly evaporated and dried. The detector was then packaged for irradiation. The samples were irradiated for 10 min in the High Flux Beam Reactor or for 150 min in the Medical Research Reactor to give, in either case, a thermal neutron fluence of 10^{17}/cm^2.

Track etching and counting

The activated detector slides were allowed to decay for 2 days, after which they could be handled in a regular fume hood because ^{31}Si, the major radioactive product (with a half-life of 2.6 hours), had completely decayed. Most of the residual β-activity originated from the plastic packaging, which was discarded. The silica slide was washed in dilute HNO_3 and rinsed in distilled water. Tracks were enlarged by etching in concentrated HF (48%) in a Teflon poly(tetrafluoroethylene) beaker, rinsed with distilled water, and air dried. Etching enlarges the tracks to about 10 μm, at which point they are large enough to be easily observed under a microscope. Etched tracks are of characteristic shape and have good contrast, as shown in Figure 2. The tracks are counted manually and can be easily distinguished from background. We estimate that the slight variations in shape and appearance make a relatively small contribution of about 3% to the overall uncertainty.

Slides were examined under a light microscope using bright-field illumination at 100× magnification and a video camera. Etched tracks are approximately 10 μm long and have good contrast. Three contiguous 1-cm^2 sections of the detector, corresponding to sample, flux monitor, and detector background areas, were delineated. ("Detector background" refers to tracks from only the detector and incidental contaminants.)

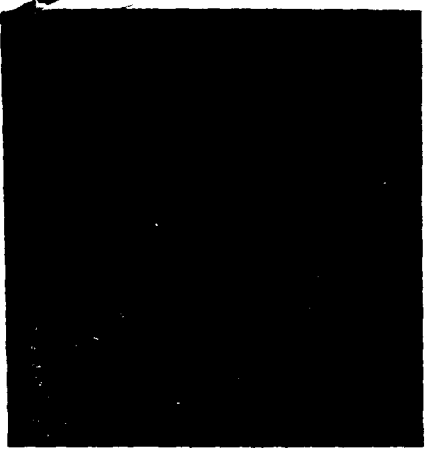
The darker circular images of the tracks illustrated in Figure 2 originate from fragments that were incident at right angles to the detector surface; the lighter oblong images derive from particles that impinged more acutely and led to a shallower and somewhat elongated track. The sensitivity of the procedure to U contamination can be put in perspective by comparing Figure 2 with Figure 3; the cluster in the latter derives from a U hot spot on the detector surface. The star pattern arises because many tracks emanate from one locus in the particle that is rich in U.

Quantitative aspects

To determine the decontamination factor for U, urine samples were spiked with 100 mg of natural U and processed through the entire experimental procedure. Eight hundred tracks were observed in the fraction corresponding to the region of Pu elution, which leads to a decontamination factor of 5 × 10^8. Because U is typically present in nanogram quantities in urine, it will contribute heavily to background unless a high degree of U decontamination is achieved.

The detector background is 4 ± 5 tracks (n = 52), and if the detection limit is taken as three times the background uncertainty, a value of 15 tracks, which corresponds to <10 aCi, results. The procedure is therefore several orders of magnitude more sensitive than α-spectroscopy, where measurements are limited to the femtocurie level. Mass spectrometry and related methods are closer in sensitivity to FTA in that attocurie sensitivity can be obtained (*7*). A drawback, however, is that memory effects will cumulatively add to background, especially for low-level determination. This situation is absent in FTA, where detectors are used only once. On the other hand, mass spectrometry allows element and isotope specificity and provides faster sample turnaround.

The absolute sensitivity (instrumental detection limit) of FTA is more than adequate for our application because reagent and urine blanks contain appreciable levels of ^{239}Pu or other inseparable contaminants. Levels in urine



blanks averaged 48 ± 34 aCi if all the tracks were attributed to ^{239}Pu. Using the detection criterion defined above, a detection limit in urine of 100 aCi or 4×10^6 atoms (above background) results.

The track count from urine blanks obtained from a Brookhaven employee exposed only to environmental levels of ^{239}Pu averaged 27 ± 19 (n = 26). These tracks do not necessarily result from ^{239}Pu; they could also be obtained from residual U. The wide variation in background is consistent with the presence of either Pu or U. Pu excretion from the body is irregular and depends on dietary and other factors (*8*); the quantity of U excreted is also variable.

Results from urine blanks spiked with various levels of ^{239}Pu are given in Table I. As expected, the relationship is linear (r^2 = 0.990):

$$\text{Track count} = 0.56\ \text{aCi}\ (^{239}\text{Pu}) - 0.60 \quad (1)$$

Each attocurie gives rise to about 2 tracks under our conditions. The data were obtained over a period of several months, and the uncertainty is long term in that it includes variations in background and recovery resulting from variable reagent purity and sample handling. The limit of quantitation—the point at which the signal is 10-fold greater than the standard deviation—is about 300 aCi.

A few hundred analyses have been performed to date, and up to 1 fCi/L of ^{239}Pu have been found in Marshallese urine samples. Representative results from split samples (each of about 500 mL) are as follows (in attocuries): 90/150; 170/250; 520/600; 130/50; 330/250; 300/210; and 230/250. The precision at or above the LOQ is adequate for meaningful dose estimates to be made. The technique is also currently being applied to weapons workers potentially exposed to Pu.

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